

10p

N65-35242

FACILITY FORM 602

(ACCESSION NUMBER)

(THRU)

(PAGES)

(CODE)

(NASA CR OR TMX OR AD NUMBER)

(CATEGORY)

THERMODYNAMICS AND APPLICATIONS OF BIOELECTROCHEMICAL
ENERGY CONVERSION SYSTEMS

BY

MICHAEL G. DEL DUCA ~~NAME~~ and JOHN M. FUSCOE

[1964] 608

GPO PRICE \$ _____

CFSTI PRICE(S) \$ _____

Hard copy (HC) 3.00

Microfiche (MF) .50

ff 653 July 65

BIOTECHNOLOGY AND HUMAN RESEARCH
OFFICE OF ADVANCED RESEARCH AND TECHNOLOGY
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION,
WASHINGTON, D. C.

Presented at the
6th AARD Combustion and Propulsion
Colloquium on Energy Sources
and Energy Conversion, Cannes, France,
March 16-20-1964

Unc. Available to ~~XXXXXX~~ and
~~XXXXXX~~

TABLE OF CONTENT

I.	Introduction -----	1
II.	Fundamental Principles -----	3
	A. Change in Free Energy and Electrical Potential ---	3
	B. Generation of Electrical Potentials in Biological Oxidation-Reduction Reactions -----	5
	1. Bioanode -----	7
	(a) Glucose-Glucose Oxidase System -----	9
	(b) Amino Acid-D-Amino Acid Oxidase System ---	11
	(c) Urea-Urease System -----	12
	2. Sulfate Biocathode -----	14
	C. Biological Generation of Fuels for Electrical Energy	14
III.	Applications of Bioelectrochemical Conversion -----	17
	A. General -----	17
	B. Sources of Power or Energy -----	18
	1. Requirements for 1-10 Milliwatts -----	18
	2. Requirements for 2-5 Watts -----	20
	3. Requirements for 20 Watts -----	22
	4. Requirements greater than 20 Watts -----	23
	(a) Urinary Waste as a Source of Energy -----	27
	(b) Vegetative Sources of Energy -----	27
	C. Detection and Generation of Control Signals -----	32




LIST OF FIGURES AND TABLES

	Page
Figure 1 -- Biological Electron Transport	6
Figure 2 -- Polarization of a Glucose Bioanode	10
Figure 3 -- Relation of Cell Current to Extent of Urea Hydrolysis	13
Figure 4 -- Laboratory Magnesium Sulfate Bio Cells	21
Figure 5 -- Yearly World Energy - Country Distribution	25
Figure 6 -- Yearly World Energy - Population Distribution	26
Figure 7 -- Anodic Polarization of Platinum in Urine	28
Figure 8 -- Current-Voltage of Vegetative Bio-Fuels	30
Figure 9 -- Current-Voltage of Biologically Catalyzed Vegetative Bio-Fuels	31
Figure 10 -- Applicable Power Levels for Bioelectrochemical Conversion	34
Table I -- Electrode Potentials of Biochemical Reaction	8
Table II -- Biochemical Generation of Fuels	16
Table III -- Yearly World Energy Distribution	24

I. Introduction

Bioelectrochemical energy conversion (i.e., converting chemical free energy of biologically catalyzed reactions to electrical energy) is not a newly discovered phenomenon. As long ago as 1786 Galvani observed that a frog muscle twitched when touched with copper-zinc couples in this early insight into the electrical characteristics of biological systems.

Over 50 years ago (1912) Potter demonstrated that a "bacterial culture during the process of energy conversion is in a sense, therefore, a primary electrical half cell and as such should conceivably be able to perform work."

Cohen, in 1930, obtained 1.25 milliamps from six yeast cells biochemically converting glucose in solution. Through this process he later built a battery able to furnish 2 milliamps at about 35 volts.

During the past 10 years interest in biochemical energy has increased fantastically, reaching exponential proportions within the last five. Particular attention in the past two years has been directed to applying the results of research and development to this type of energy

conversion. This paper will consider these applications in a state-of-the-art review and will present a synopsis of suggested applications ranging from the use of bioelectric currents to identify toxic materials and power human implanted cardiac pace makers to the generation of electric power in remote areas of the world.

Research in biochemical energy conversion -- limited to the laboratory -- is in its infancy. Available data are inadequate to form a sound basis for defining specific engineering and economic criteria that might point the way toward selecting particular technology for development and later applications. Since criteria for bioelectrochemical systems cannot now be defined, it is strongly suggested that present and extrapolated characteristics of such systems should not be compared with those of other energy converters in use or being developed. Such a practice could be discouraged and might result in failure to realize the full potential of bioelectrochemical conversion.

II. Fundamental Principles

The fundamental principles of bioelectrochemistry should be considered in order to fully realize the potentialities of this field. Some insight into the electrical potentials associated with bioelectrical-chemical systems may be derived from the Second Law of Thermodynamics. This law determines the relation between free energy changes and the standard oxidation potential of reactions occurring in biological systems.

A. Change in Free Energy and Electrical Potentials

As is well known, the free energy F , of the reactants and products in the biochemical system $aA + bB \rightleftharpoons cC + dD + \dots$ may be expressed in terms of the chemical potentials as

$$F_{\text{reat}} = a\mu_A + b\mu_B + \dots \quad (1)$$

$$F_{\text{prod}} = c\mu_C + d\mu_D + \dots \quad (2)$$

At constant temperature and pressure, the free energy change of the reaction is given by

$$\begin{aligned} \Delta F_{TP} = & c\mu_C + d\mu_D + \dots \\ & - a\mu_A - b\mu_B - \dots \end{aligned} \quad (3)$$

The chemical potential μ is given in the standard state μ_0

$$\mu = \mu_0 + RT \ln \bar{a} \quad (4)$$

where \bar{a} is the activity of the species in question under defined conditions. Substitution of (4) and (3), and collecting terms,

$$\Delta F_{TP} = \Delta F_T^\circ + RT \ln \frac{\bar{a}_C^c \cdot \bar{a}_D^d}{\bar{a}_A^a \cdot \bar{a}_B^b} \quad (5)$$

When the reactants are incorporated in an electrochemical conversion system.

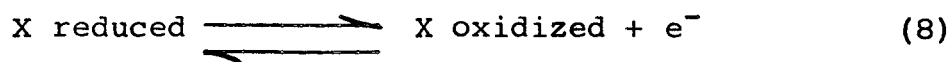
$$-\Delta F = nFE \quad (6)$$

where F is the Faraday constant, E the electrode potential and n is the number of electrons associated with the reaction. Equation (6) is of great importance since it permits the calculation of standard electrode potentials E_0 (standard conditions and unit activity) from free energy data. Substituting $-nFE$ for ΔF and nFE_0 for ΔF° and substituting concentration (C) for activities (although an approximation).

Equation 5 becomes

$$E = E_0 - \frac{RT}{nF} \ln \frac{C_C^c \cdot C_C^d}{C_A^a \cdot C_B^b} \quad (7)$$

from the standard oxidation - reduction reaction



$$E = E_0 - \frac{RT}{nF} \ln \frac{X_{ox}}{X_{red}} \quad (9)$$

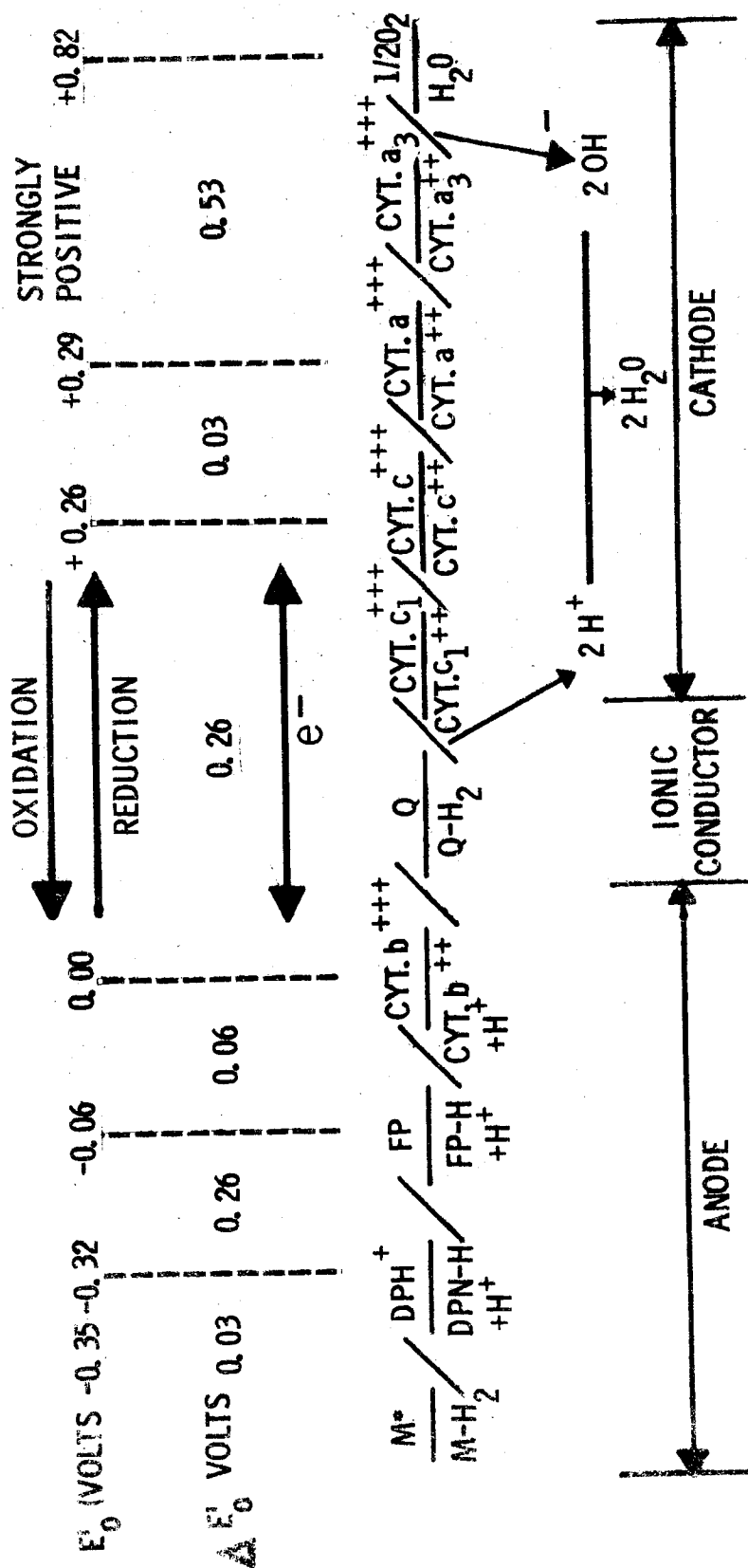
B. Generation of Electrical Potentials in Biological Oxidation--Reduction Reactions

Energy transfer reactions in biological systems are of the oxidation-reduction type. Equation (9) permits the calculation of the electrical potential of these systems for various degrees of oxidation.

In general there are two main schemes for the biological oxidation of organic materials. The first (fig. 1) is based on an initial hydrogen removal from the reduced (or fuel) molecule, followed by a successive series of hydrogen atom and electron transfers along a chain of redox couples resulting in electron transfer to oxygen. The entire process is catalyzed by biological catalysts (enzymes).

The second involves the direct activation of molecular oxygen by the oxidase enzymes. Three types of oxidases are those which catalyze: (1) direct oxygen addition to a molecule, (2) reduction of one oxygen atom

BIOLOGICAL ELECTRON TRANSPORT



M- METABOLITE
 DPN - DIPHOSPHO PYRIDINE NUCLEOTIDE
 FP - FLAVOPROTEIN
 CYT - CYTOCHROME
 Q - COENZYME Q

(in O_2) by electron transfer and direct addition of the other to a reacting molecule, and (3) reduction of oxygen to H_2O_2 or H_2O .

In the work reported to date on application of the foregoing principles to bioelectrochemical cells the procedure has been to break the oxidation sequence so that the entire enzyme system is in one or the other of the electrode compartments. Systems such as those shown in figure 1 are divided into an oxygen electrode and an organic fuel-enzyme system separated by a suitable ionic conductor. Of particular significance is the step in the oxidation sequence at which electron transfer to an inert electrode can occur.

1. Bioanodes

Table I lists a number of potential bioelectrochemical oxidation-reduction couples which are anodic to oxygen at the more normal biological pH of 7. The potentials in Table I indicate that with the appropriate enzyme catalysts, substances found in natural sources such as acetaldehyde, xanthine, glucose, and cysteine may be feasible as fuel electrodes with an

TABLE I

ELECTRODE POTENTIALS OF BIOCHEMICAL REACTIONS

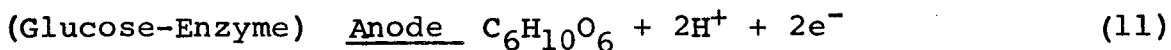
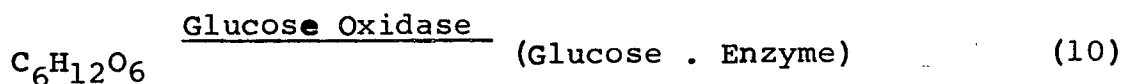
<u>Electrode Couple</u>	<u>Enzyme</u>	<u>E*_{m7}</u>
Acetate/Acetaldehyde	Xanthine Oxidase	-0.58
Latic Acid/Xanthine	Xanthine Oxidase	-0.39
Gluconolactone/Glucose	Glucose oxidase	-0.36
Cystine/Cysteine	None	-0.33
Acetaldehyde/Ethanol	Alcohol dehydrogenase	-0.20
Pyruvate/Lactate	Lactic dehydrogenase	-0.18
Oxaloacetate/Malate	Malic dehydrogenase	-0.16
Fumarate/Succinate	Succinic dehydrogenase	+0.02
Dehydroascorbate Ascorbate	Ascorbic oxidase	+0.06
Ferricytochrome c/Ferrocyclochrome c		+0.27
Oxidized cyt. oxidase/Cytochrome oxidase		+0.29
$O_2 + 4H^+ + 4e \rightleftharpoons 2H_2O$		+0.82

*E_{m7} is the potential at pH 7 in the presence of equal concentrations of oxidized and reduced forms of reactants.

oxygen (air) cathode. The energy density of these materials is on the order of 5 pounds per kilowatt-hour. Several enzymes catalyzed and whole organism-catalyzed redox couples have been studied under laboratory conditions. The results are discussed below:

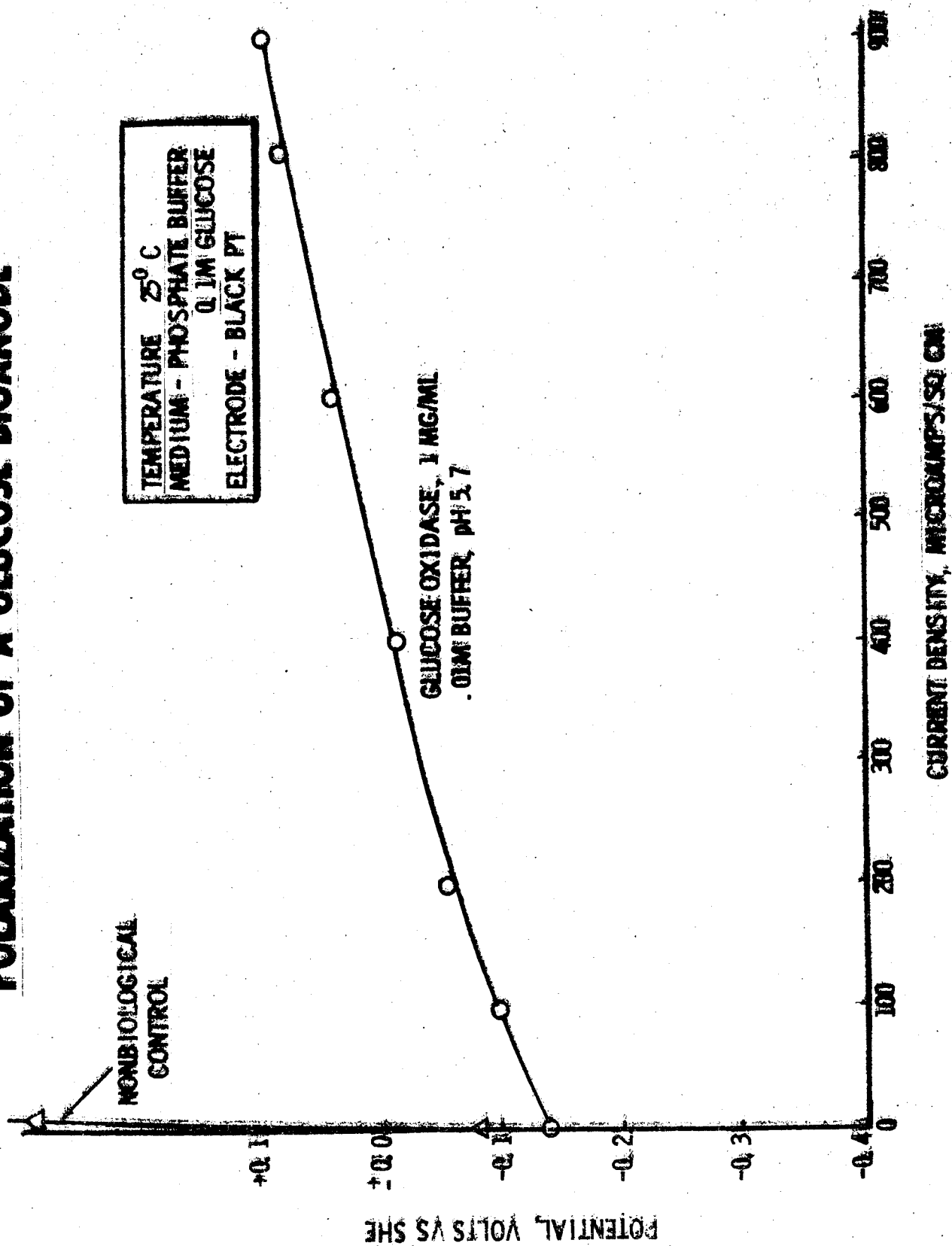
(a) Glucose - Glucose Oxidase System

The electrode mechanism has not been critically described for this system. Several mechanisms are suggested. In a direct mechanism glucose and the enzyme glucose oxidase react to form a complex intermediate. The intermediate is oxidized at the anode to gluconolactone and the enzyme is liberated. (Equations 10 and 11).



Anodic polarization data for the glucose-glucose oxidase bio-anodes is shown as figure 2. In these studies, the enzyme glucose oxidase is incorporated in the pt-black electrode surface and glucose (reduced form is added to the cell). As shown in figure 2, the electrochemical

POLARIZATION OF A GLUCOSE BIOANODE



oxidation of glucose on platinum is markedly enhanced by the incorporation of the glucose oxidase as a catalyst.

(b) Amino Acid - D - Amino Acid Oxidase
System

The normal aerobic reaction products of the amino acid D - amino acid oxidase (DAO) system include the pyruvic acid derivative of the amino acid, hydrogen peroxide and ammonia.

Experiments have been conducted on the bioelectrochemical behavior of the three amino acids Tryptophane, tyrosine and phenylalanine and their respective D-amino acid oxidase reaction products indole-3-pyruvic acid, para-hydroxy phenyl pyruvic acid, and phenyl pyruvic acid. Results to date on platinum electrodes show that observed currents are derived from the electrochemical oxidation of the aromatic pyruvic reaction products. Ammonia not peroxide contributed appreciably to the electrical currents. Currents of the order of $350 \mu\text{A}/\text{cm}^2$ at 200 mv have been derived with indole-3-pyruvic acid.

Currents on the order of $45 \mu\text{A}/\text{cm}^2$ at 200 mv have been derived from cells in which indole pyruvic acid is generated enzymatically from Tryptophane.

These studies indicate that: (1) the current limitation is enzymatic, (2) the reactive species is the enol form of the keto acid derivative of the naturally occurring amino acids (absorption spectra data), and (3) further studies are needed to determine optimal condition for enzyme and electrochemical activity in order that the potential of utilizing complex natural occurring materials as electrode reactants may be exploited.

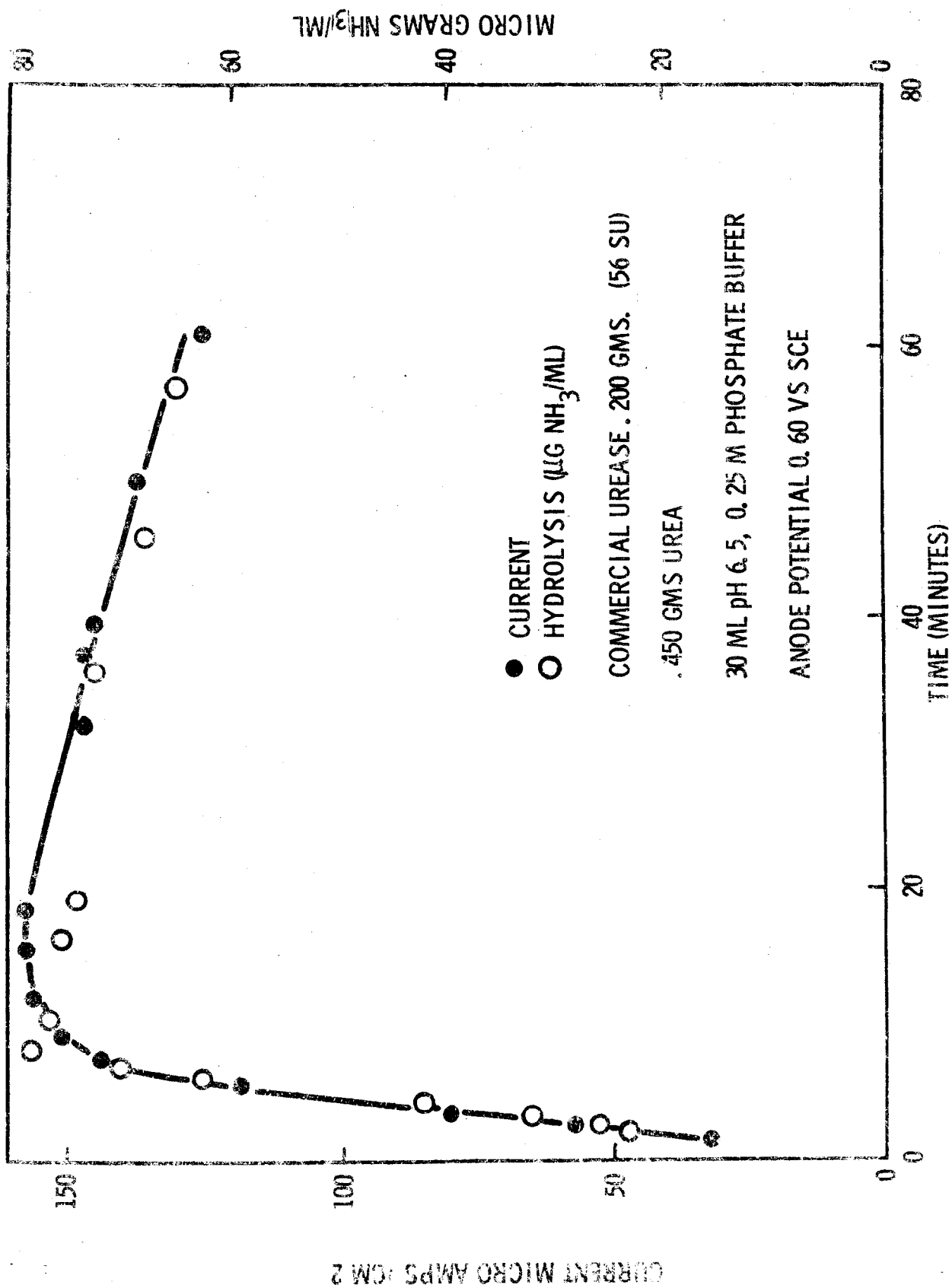
(c) The Urea-Urease System

Theoretically it would be expected that in this cell the enzyme urease would generate ammonia from urea and the ammonia electrochemically oxidized:



In studies to date the Urea-Urease System has presented a rather consistent dilemma. In cells with bright platinum electrodes in 0.25 M ammonium nitrate in Tris buffer (tris-hydroxymethyl amino methane) at pH of 6.5 - 9.0 ammonia was reportedly not oxidized. In the same cell, nevertheless, the action of urease resulted in the augmentation of the cell current and the enzymatic hydrolysis of urea parallels the cell current, (fig. 3). Other studies have shown that

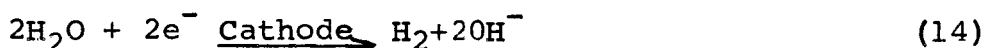
RELATION OF CELL CURRENT TO EXTENT OF UREA HYDROLYSIS



at pH 6 in a K Cl - citrate buffer using platinum impregnated carbon electrodes incorporating the urease enzyme, short circuit currents of 1-3 ma/cm² were derived on addition of urea. Further studies on this system are being conducted.

2. Sulfate Biocathode

Potentials associated with biologically catalyzed reductions have also been investigated. The reduction of sulfate to hydrogen sulfide by the organism Desulfovibrio desulfuricans, attached to porous iron electrodes, has been extensively studied. However, the mechanism for this electrode has not been completely established. It is thought that the reactions for the bioelectrochemical reduction of sulfate may be as follows:



C. Biological Generation of Fuels for Electrical Energy

Biological energy sources may also indirectly generate electricity. Biochemical agents may generate chemical species specifically tailored to a biological energy

conversion system optimized to the requirements and consistent with available fuel. Examples include the generation of hydrogen, ammonia, or methanol from higher molecular weight materials (such as sugars, proteins, fats, starches, urea) by means of whole living cells or cell free extracts or crystalized enzymes.

Samples of biochemical systems for the generation of primary fuels from complex materials are presented as Table II. The literature has been reviewed and over 200 references are included in the bibliography under "Generation of Bioelectrochemical Fuels."

Applications of bioelectrochemical conversion are discussed in the following paragraphs.

BIOCHEMICAL GENERATION OF FUELS

NATURAL FUEL	NATURAL SOURCE	CATALYST	PRIMARY FUEL	LB/KWH
AMMONIA GENERATORS				
UREA	URINE	UREASE (CRYSTALLINE ENZYME)	NH ₃	.83
AMINO ACIDS	PROTEIN DIGESTS	AMINO ACID OXIDASE (ENZYME)	NH ₃	1.2-3
HYDROGEN GENERATORS				
FORMIC ACID		FORMIC DEHYDROGENASE (ENZYME)	H ₂	1.8
METHANE GENERATORS				
PROPRIONIC ACID	FATS, OILS	METHANOBACTERIUM PROPRIONICUM (LIVING BACTERIA)	CH ₄	1.0

III. Applications of Bioelectrochemical Conversion

A. General

Research and development looking toward the applications of bioelectrochemical conversion is in its early phases. However, at present four general areas for potential future applications appear to be emerging.

These include:

- Power sources for supplying relatively small amounts of electrical energy in specialized applications and locations.
- Detectors for specific contaminants in low concentrations.
- Sensors for generation of control signals.
- Catalysts for the generation of electrochemical reactants from complex naturally occurring materials and from waste materials.

Available information indicates that bioelectrochemical power generation will most likely be limited to specialized applications. Competition with major power generation methods is unlikely. Bioelectrochemical power

generation will probably supply limited emergency power, serve remote and unattended power supplies, and supplement existing natural fuels (such as vegetation) when transportation costs for additional fuels are excessive.

B. Sources of Energy or Power

As power sources bioelectrochemical converters may be arbitrarily classified in terms of power levels for suggested applications.

1. Requirement for 1-10 milliwatts

Low power level implantable transducers, stimulators, and other electronic devices are needed to regulate physiological and biological functions.

For example, the present day peacemaker used in the regulation of the electrical profile of the human heart requires electrical energy at about 2.4 volts with current pulses 2 milliseconds in duration at a frequency of one to two/second. The average current and power are about 25 μ amperes and 100 μ watts, respectively. This energy is normally supplied by five primary cells, calculated to last for two to three years, which are made part of the implantable package. Surgery is required to replace the

dead batteries. The operating life of batteries for other implantable devices may be much less.

The development of implantable energy converters which derive their energy on a continuous or periodic basis from physiological fluids may be quite advantageous. Consequently, the development of an implantable fuel cell with output of 200 μ watts at a voltage level in the vicinity of 1 volt may be a very worthwhile objective.

The literature available on the measurement of oxidation-reduction potentials of mammalian blood is extensive. Electrode potentials ranging from 0.5 to +0.3 are relatively common in mammalian body fluids. With a fuel cell of 0.5 volt at a current density of 10 μ amperes per square centimeter, the power output would be 5 μ watts per cm^2 of electrode surface area. Even at such low current densities the peacemaker power requirements could easily be satisfied with electrode surface areas of the order of 100 cm^2 .

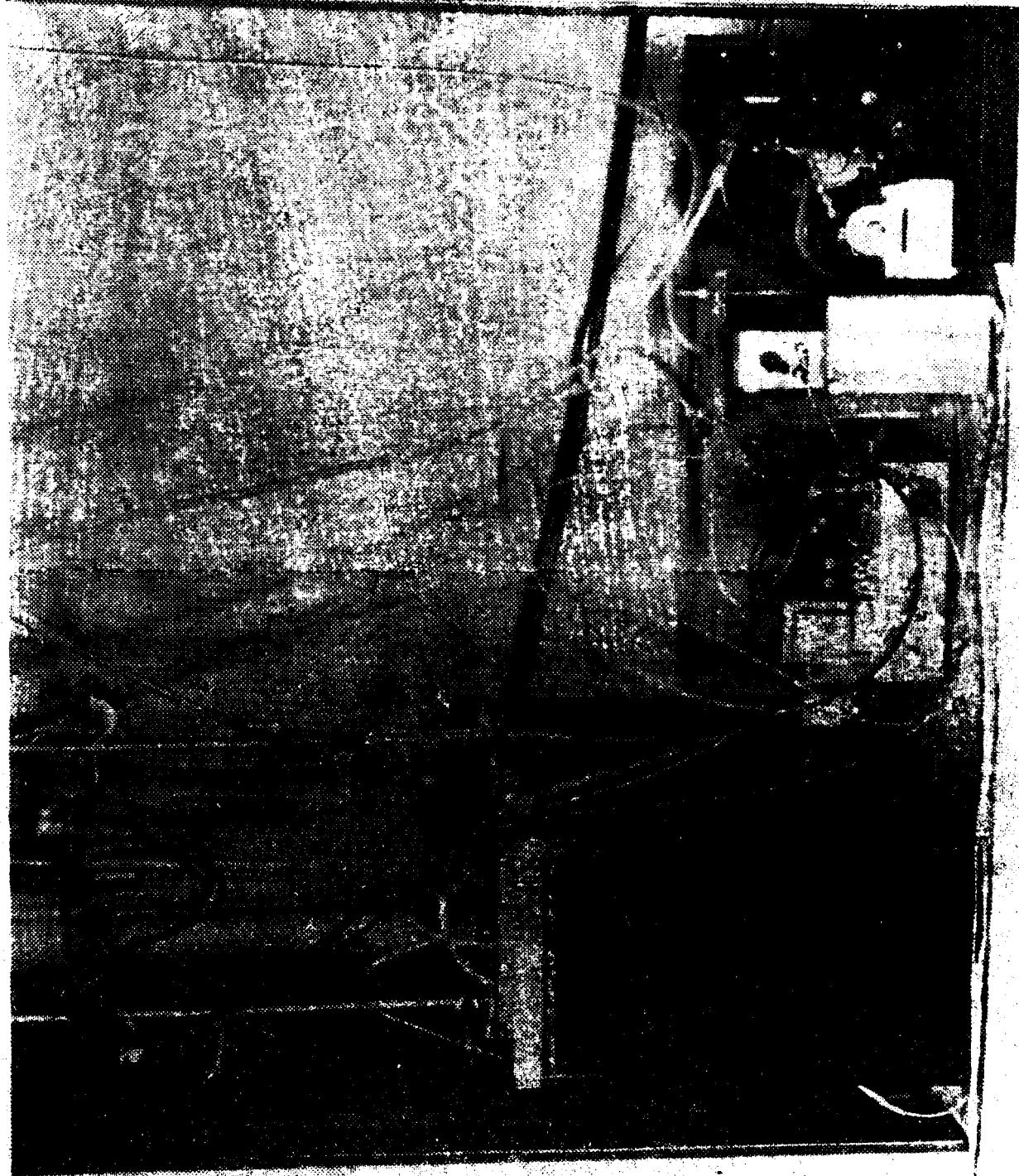
In an experimental analog of the conditions found in the blood stream the open circuit voltage was

approximately 0.1 volt when platinum black electrodes were used. Current densities ranging from 10 to 50 μ amps per square centimeter were found to be linear with the polarization voltages having a slope of nearly 2 1/2 millivolts per μ ampere per square centimeter.

2. Requirements for 2-5 watts

Attention has been given to the application of direct bioelectrochemical converters for long duration unattended power generation. Use of these converter systems to supply 2-5 watts of power in the ocean has been broadly explored. Sulfate, available in the ocean, has been studied extensively as the cathodic reactant of such a power source device. In this device bacteria (Desulfovibrio desulfuricans) are applied to the cathode where they consume hydrogen and reduce sulfate ion to sulfide ion. The anodic process is the oxidation of magnesium. Limiting current densities as high as 3 ma/cm² at -0.8V vs SCE have been reported under optimum conditions. Cell design has not been optimized but power densities in the range of 5-20 W/ft³ and energy densities of approximately 30 KWH/ft³ may be estimated for a cell to operate two years without attention. A laboratory fuel cell is shown in figure 4.

LABORATORY MAGNESIUM SULFATE BIO CELLS



3. Requirements for 20 watts

The availability of used oxygen and the enzyme urease has suggested their use in an open-cycle power generator. Several investigations have been conducted of the electrochemical performance of a urease urea oxygen (air) battery. In the future this type of battery might be used to produce relatively low power (below 20 watts) for short periods of time (2 weeks, for example).

As anode materials platinum black electro-deposited platinum, platinum impregnated carbon, active impregnated carbon, activated carbon, Raney silver, and electrodeposited nickel have been investigated. Crystalline enzyme catalysts would be incorporated in the electrodes; monel screens and porous carbon blocks used as current collectors. A 3 M KCl, citrate buffer at pH6, resulted in maximum limiting current densities. The air cathode is a porous platinized carbon electrode. These fuel cells have achieved an open circuit voltage of about 0.8V and a short circuit density of 3.6 amp/ft² for short periods of time.

4. Power Requirements Greater than 20 watts

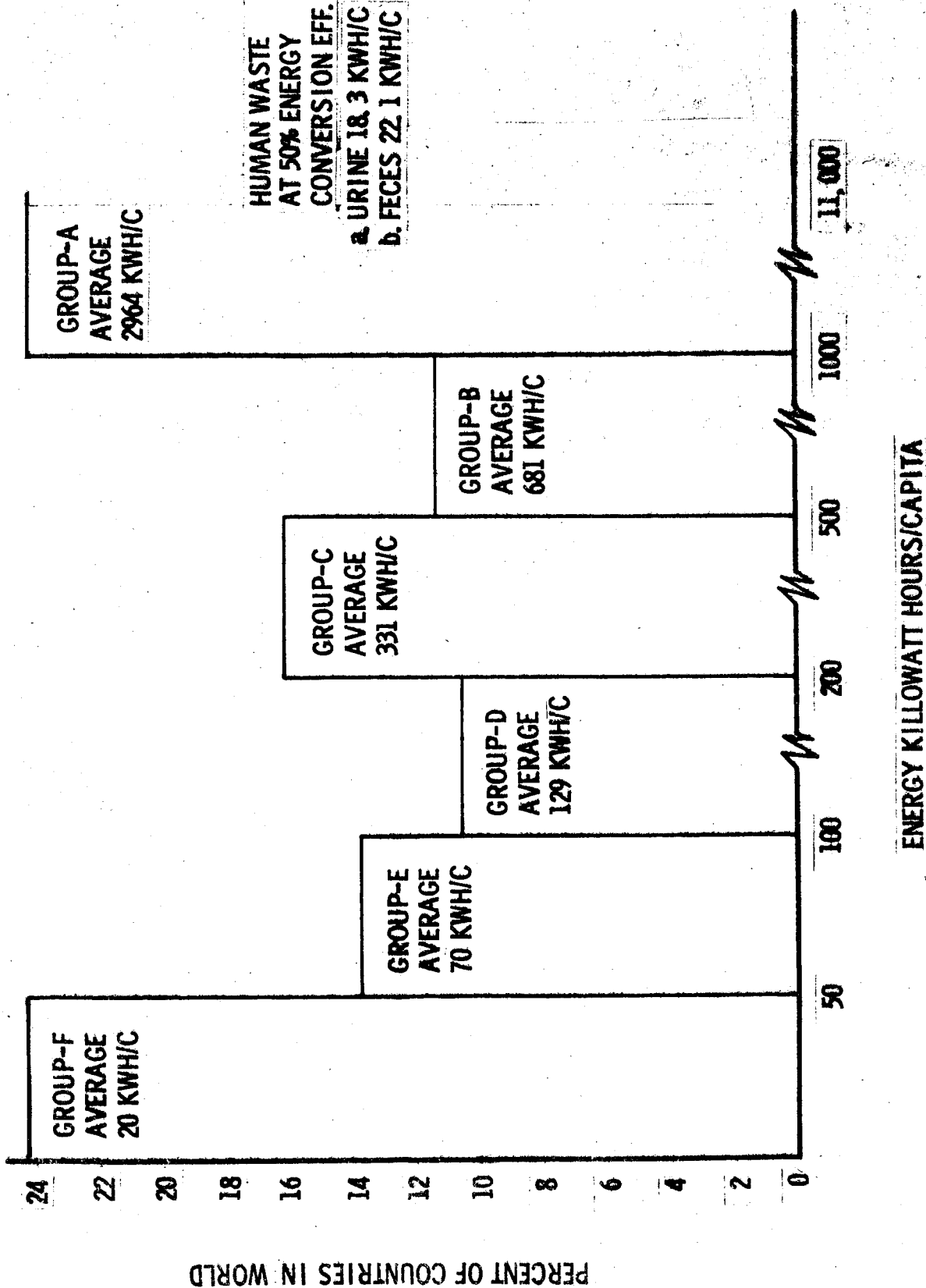
Several factors need to be analyzed when considering bioelectrochemical convertors for power applications in excess of 20 watts. Worldwide energy demand and fuel logistics are two of these important considerations. A distribution of the world's yearly electrical energy consumption is shown as table III, distribution by country as figure 5, and distribution on the basis of world population as figure 6. These data show that about 24 percent of the world's countries, with 37 percent of its population, have an average yearly electrical energy consumption of only 20 kilowatt hours per capita. It is evident from this fact that special remote locations throughout the world may well be provided with a minimal power source at minimal costs by using biological sources of energy.

Biological sources of energy could be used for the generation of electrical power directly by bioelectrochemical energy conversion or indirectly by biocatalytic generation of simple fuels from complex

TABLE III

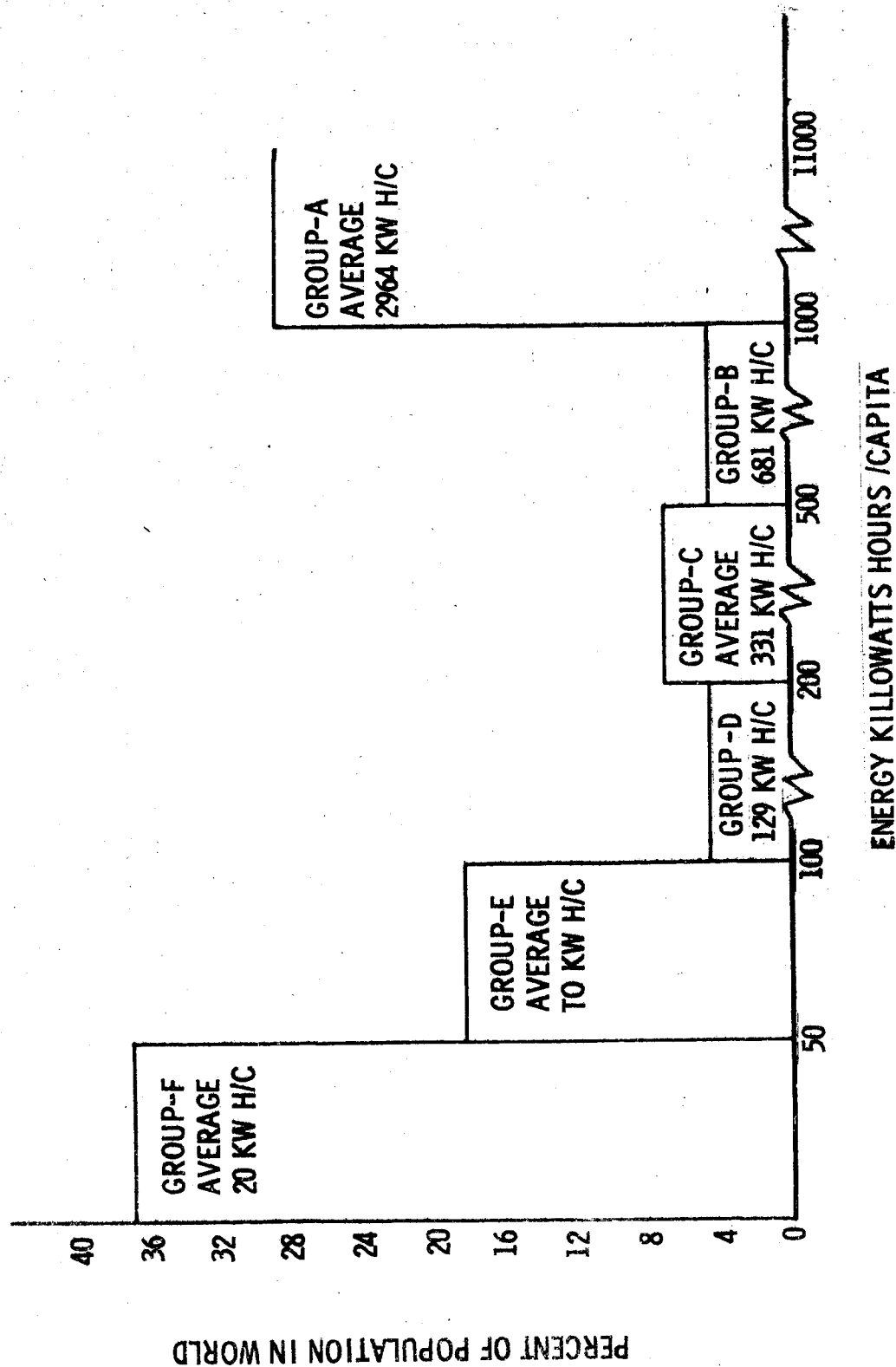
<u>Group</u>	<u>Range KWH per capita</u>	<u>Average KWH per capita</u>	<u>Percent Countries</u>	<u>Percent Population</u>
A	1,000-10,346	2,964	24.2	28.9
B	500-1,000	681	11.3	4.3
C	200-500	331	16.1	6.9
D	100-200	129	10.5	4.4
E	50-100	70	13.7	18.3
F	0-50	20	24.2	37.2

YEARLY WORLD ENERGY-COUNTRY DISTRIBUTION



YEARLY WORLD ENERGY-POPULATION

DISTRIBUTION



naturally occurring materials. Of particular significance is the utilization of biological energy sources in their natural form.

(a) Urinary Waste as a Source of Energy

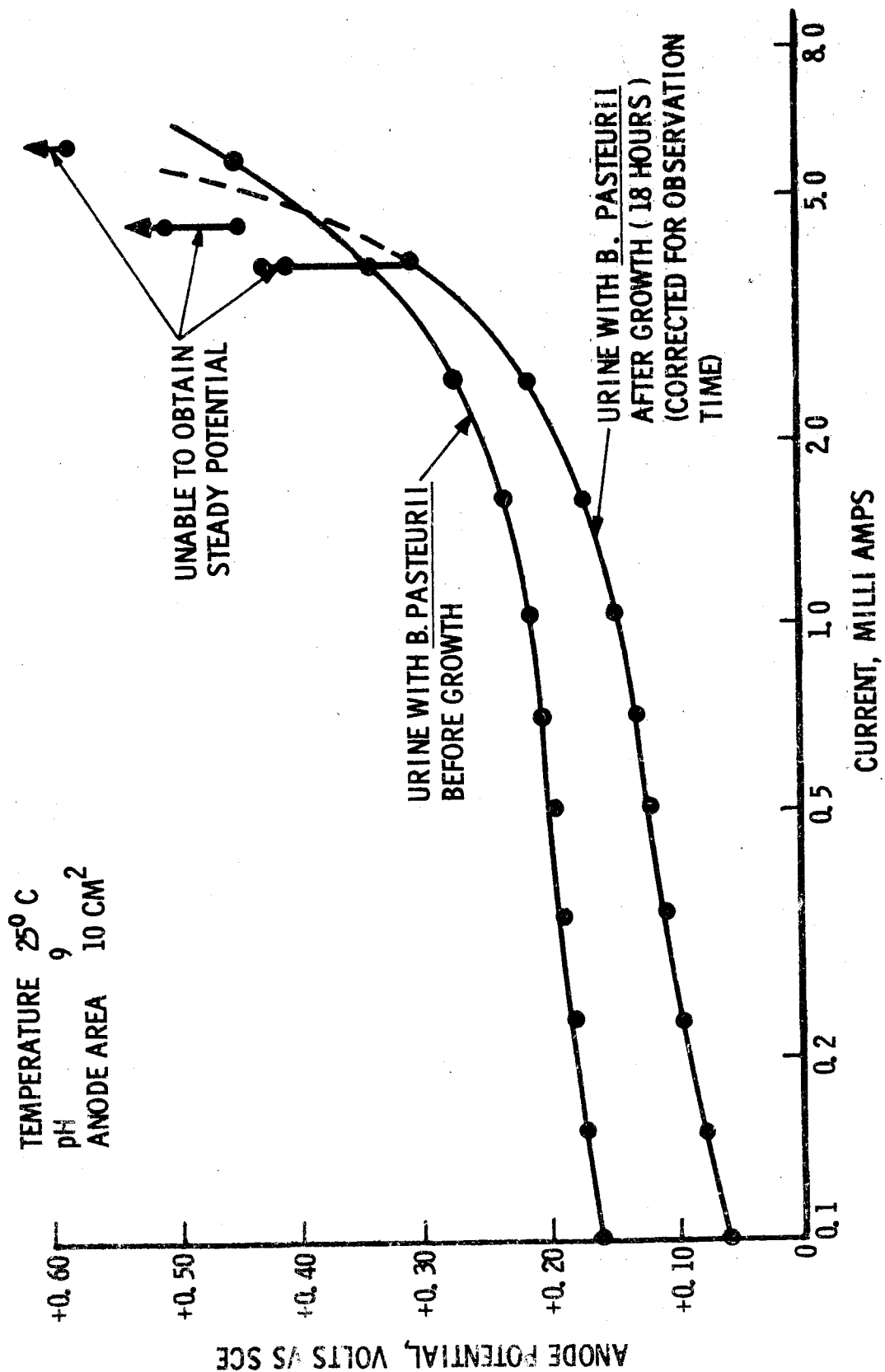
Theoretically, bioelectrochemical conversion of urea in human urinary wastes could supply on the order of 18 KWH per capita yearly based on a conversion efficiency of 50 percent.

Work has already been conducted on the electrochemical behavior of systems fueled by raw urine and catalyzed by growing bacteria. The bacteria Bacillus pasteurii has been studied in a system because of its established ureolytic activity. Preliminary data (fig. 7) indicate that a preliminary 18-hour incubation of raw urine with this bacteria provides increased current. At 170 mV, for example, the current carrying capacity of the electrode fueled by preincubated material was greater by a factor of 15 than the unincubated material.

(b) Vegetative Sources of Energy

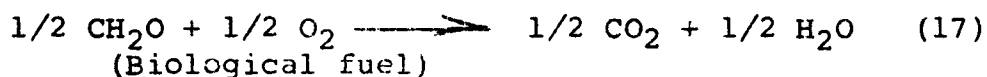
Several other systems using natural materials as fuels have been studied. Laboratory studies have shown

ANODIC POLARIZATION OF PLATINUM IN URINE, EFFECT OF *B. PASTEURII* GROWTH



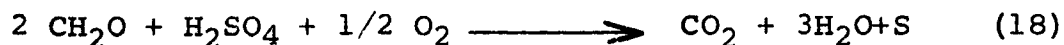
that some power gains may be realized by the biological catalyzation of electrochemical systems using natural materials such as fresh mushrooms, toadstools, sucrose, and algae as fuels and oxygen (air) cathodes.

Chemical digestion of natural materials with sulfuric acid and potassium hydroxide results in the generation of electrochemically active organic and inorganic material and an over-all net reaction may be written:



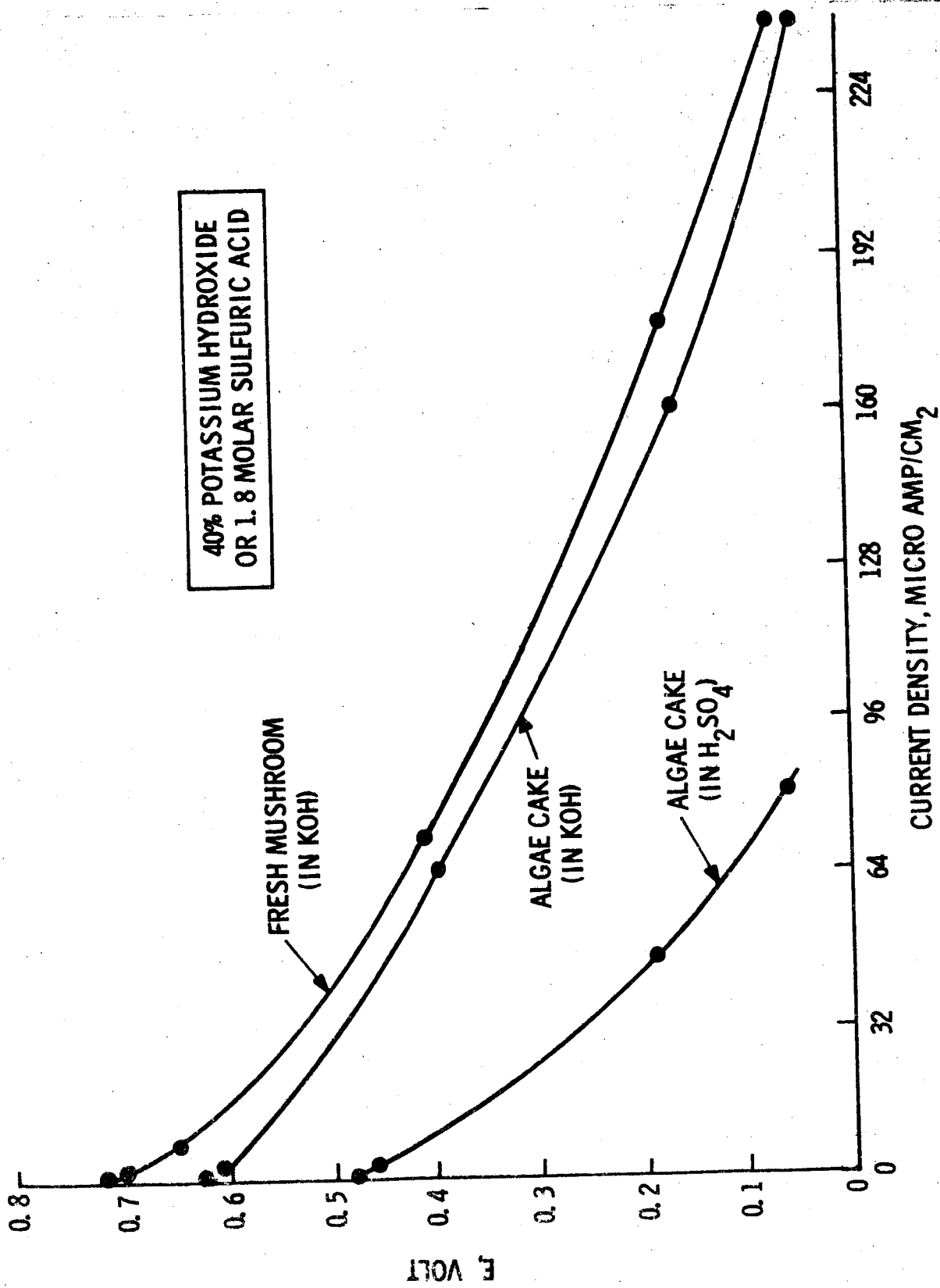
Some laboratory results for the non-biologically catalyzed systems are presented as figure 8.

The biological catalyzed oxidation of natural fuels with sulfuric acid using the bacteria Desulfovibrio may be used as anodic reactants in a system with an oxygen (air) cathode. The over-all net reaction may be given as follows:

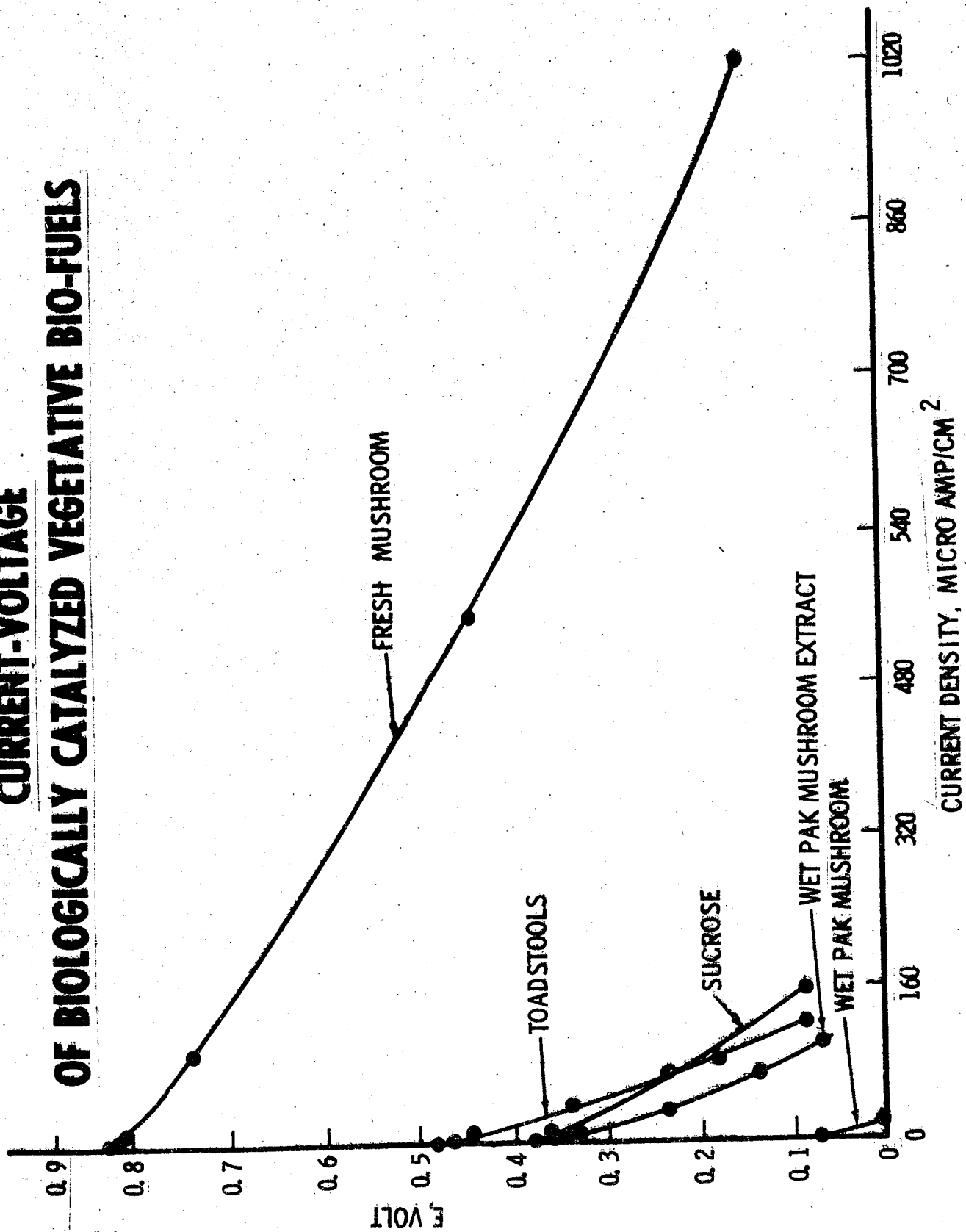


Laboratory results for several systems studied are presented as figure 9. A comparison of

CURRENT-VOLTAGE OF VEGETATIVE BIO-FUELS



CURRENT-VOLTAGE OF BIOLOGICALLY CATALYZED VEGETATIVE BIO-FUELS



figures 8 and 9 indicates a greater than 4 fold increase in current densities may be realized by biological catalysts in systems using complex natural fuels. These studies have been conducted under laboratory conditions in order to elucidate the parameters effecting bio-electrochemical activity. Much work remains to be done before the feasibility is established of utilizing such phenomena as practical applicants where complex natural materials are readily available.

C. Detection and Generation of Control Signals

Another area of applications for bioelectrochemical conversion is in (1) the detection of simple and complex chemical species in extremely low concentrations, and (2) in the use of electric signals associated with biochemical systems for control. Illustrative work to date includes the toxic gas detector and myoelectric serve boost system.

The influence of various chemicals on the activity of naturally occurring microorganisms has been utilized in the detection of toxic agents.

The activity of the microorganism has been found to be proportional to the concentration of various agent.

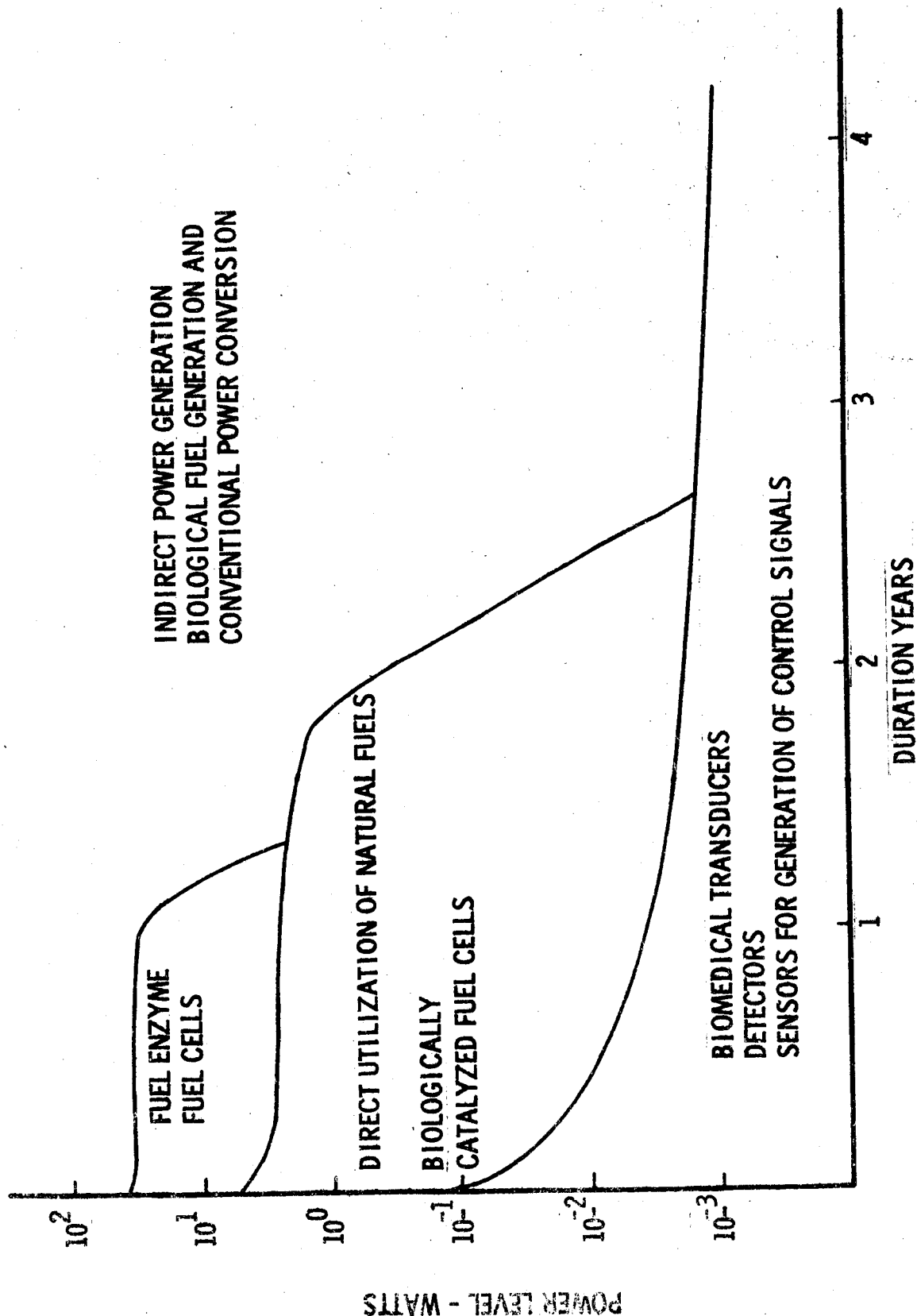
Specific biological agents have been preselected which will provide detection of trace materials at concentrations of parts per billion.

The feasibility has been demonstrated of using electrical signals associated with contraction of muscles to control a servo boost system which would enable an individual to remotely handle adverse tasks. In this application silver foil electrodes are attached externally to a subject and a potential of 1-3 millivolts peak to peak at 3-1000 cycles/second are picked off and used to direct an external control logic.

Finally, in the broadest sense, bioelectrochemical energy conversion may show numerous and varied applications. Such applications are arbitrarily organized according to the level of associated power as figure 10. The applications illustrated do not represent a complete survey, since the entire area is relatively new and a complete picture has not been developed.

However, it is hoped that the above discussion will serve to stimulate thinking in this area.

APPLICABLE POWER LEVELS FOR BIOELECTROCHEMICAL CONVERSION



POWER LEVEL - WATTS

REFERENCES

- [1] ALBERT, A. (1963). Nonlinear Regression Made Computationally Easy. Report, The ARCON Corp., Lexington, Mass.
- [2] CHUNG, K. L. (1954). On a Stochastic Approximation Method. Ann. Math. Statist. 25 463-483.
- [3] CRAMER, H. (1946). Mathematical Methods of Statistics, Princeton Univ. Press, Princeton. 67.
- [4] DAVENPORT, W. and ROOT, W. (1958). Random Signals and Noise. McGraw Hill, New York, 240-242.
- [5] GARDNER, L. (May 1964) Private communication.
- [6] HELSTROM, C. R. (1960). Statistical Theory of Signal Detection, Pergamon, Oxford, Chap. 11.
- [7] HELSTROM, C. R. op. cit, 312-315.
- [8] KIEFER, J. and WOLFOWITZ, J. (1952). Stochastic Estimation of the Maximum of a Regression Function. Ann. Math. Statist. 23 462-466.
- [9] PRICE, R. (1956). Optimum Detection of Random Signals in Noise with Application to Scatter-Multipath Communication. Trans. I.R.E. IT-2, No. 4, 125.
- [10] ROBBINS, H. and MUNRO, S. (1951). A Stochastic Approximation Method. Ann. Math. Statist. 22 400-407.
- [11] SACKS, J. (1958). Asymptotic Distribution of Stochastic Approximation Procedures. Ann. Math. Statist. 29 373-405.
- [12] ZADEH, L. and RAGAZZINI, J. (1950). An Extension of Wiener's Theory of Prediction, J. Appl. Phys. 21 645-655.

BIBLIOGRAPHY

The following bibliography on bioelectrochemical energy conversion and related subjects has been compiled from referenced works in the area of bioelectrochemistry. The listing is not complete. It is intended to establish a representative compilation of references needed by an investigator to embark upon research in this area. The following abbreviations were used in the bibliography to assist in the location of reports and documents.

- N numbers refer to Scientific and Technical Aerospace Reports (STAR)
- A numbers refer to International Aerospace Abstracts
- AD numbers refer to Armed Services Technical Information Agency (ASTIA) now called Defense Documentation Center (DDC)

BIBLIOGRAPHY OF BIOELECTROCHEMICAL PROCESSES

TABLE OF CONTENTS

	Page
I. General -----	1
II. Sources and Generation of Bioelectrochemical Reactants -----	1
A. Living Organisms -----	1
1. Sewage Digestion -----	1
2. Production of Ammonia and Hydrogen ---	4
B. Enzymes -----	5
1. Cellulases -----	5
2. Pectinesterases -----	7
3. Polygalacturases -----	7
4. Lipases -----	8
5. Papains -----	9
6. Ureases -----	9
7. Sulfate Reductases -----	10
8. Hydrogenases -----	11
9. Catalases -----	12
10. Role of Metal Ions in Enzyme Systems -	12

BIBLIOGRAPHY OF BIOELECTROCHEMICAL PROCESSES

TABLE OF CONTENTS

(CONTINUED)

	Page
III. Bioelectrochemical Converters and Related Studies -----	12
A. Fundamental Investigations -----	12
1. Electrode Studies -----	12
2. Bioelectric Activity -----	16
3. Organic Membranes -----	18
B. Bioelectrochemical Fuel Cells -----	20

A BIBLIOGRAPHY OF BIOELECTROCHEMICAL PROCESSES

I. GENERAL

- * Anonymous, Standard Methods for the Examination of Water and Wastewater, published jointly by the American Public Health Assn., American Water Works Assn., and Water Pollution Control Federation (1961)
- * Lloyd L. Ingraham, Biochemical Mechanisms, John Wiley and Sons, Inc., p 77 (1962)
- * Spector, W. S., Handbook of Biological Data, WADC Technical Report 56-273, ASTIA Document No. AD 110 501 (1956)

II. SOURCES AND GENERATION OF BIOELECTROCHEMICAL REACTANTS

IIA LIVING ORGANISMS

IIA1 SEWAGE DIGESTION

- * Bogen, R. H. and D. D. Chapman, Developments in Industrial Microbiology, 3, 45, Plenum Press, N.Y. (1962)
- * Brisou, B., Bull. Soc. Pathol. Exot., 54, 746 (1961)
- * Buck, T. C., C. E. Keefer and H. Hatch, Sew. and Ind. Wastes, 25, 993, (1953)
- * Buck, T. C., C. E. Keefer and H. Hatch, Sew. and Ind. Wastes, 26, 164 (1954)
- * Buck, T. C. and C. E. Keefer, Sew. and Ind. Wastes, 31, 1267 (1959)
- * Buswell, A. M., Spetic Tank to Controlled Digestion, in Biological Treatment of Sewage and Industrial Wastes, Vol.2, Eds, J. McCabe and W.W. Eckenfelder, Jr., Reinhold Pub. Corp., N.Y. (1958), 3-8
- * Buthaux, R. and D. A. A. Mossel, J. Appl. Bacteriol., 24, 353 (1961)
- * Butterfield, C. Publ. Health Repts., 50, 571 (1935)

- * Chapman, D. D., Proceedings of the Institute of Environmental Sciences, p. 283, (1960)
- * Collee, L. G., J. A. Knowlden, and B. C. Hobbs, J. Appl. Bacteriol., 24, 326(1961)
- * Consolazio, D. F., Johnson, R. E., and Pecora, L. J., Physiological Measurements of Metabolic Functions of Man, McGraw-Hill Book Company, New York (1963)
- * Eckenfelder, W. W. Jr., and O'Connor, D. J., Anaerobic Biological Treatment Processes, in Biological Waste Treatment, Pergamon Press, New York (1961), 248-269
- * Gall, L. S., Fenzer, D. B., and Helvey, W. M., Bio-Ecology of Digestion, in Biologistics for Space Systems Symposium (May 1962), Technical Documentary Report No. AMRL-TDR-62-116 (October 1962), 382-390
- * Garrett, Jr., M. T., Sew. and Ind. Waste, 30, 253 (1958)
- * Goldblith, S. A., and Wick, E. L., Analysis of Human Fecal Components and Study of Methods for Their Recovery in Space Systems, ASD Technical Report 61-419, Wright-Patterson Air Force Base, Ohio (1961)
- * Goldblith, S. A. and E. L. Wick, Contract AF 33(616)-6136, ASD-TR-61-419 (1961)
- * Goldblith, S. A. and E. L. Wick, Analysis of Human Fecal Components and Study of Methods for Their Recovery in Space Systems, U.S.A.F. Aeronaut Systems Div. Tech. Report 61-419, 1-57, (1961)
- * Golueke, C. G., W. J. Oswald and P. H. McGanky, Sew. and Ind. Waste, 31, 1125 (1959)
- * Hawkes, H. A., Ecology of Activated Sludge and Bacteria Beds, in Waste Treatment, Ed., P.C.G. Issac, Pergamon Press, Oxford, (1960), 52-98
- * Hogan, R. H., Chapman, D. D., and Ericsson, L. H., Aerobic Biological Degradation of Human Waste in Closed Systems, Advances in Astronautical Sciences, 6 (1961) 390-398
- * Ingram, W. T., TDR #AMRL-TDR-62-126, Contract AF 33(616)-7827, (1962)

- * Kaplovsky, A. J., Appl. Microbiol., 5, 175 (1957)
- * Kountz, R. R. and C. Forney, Jr., Sew. and Ind. Waste, 31, 819 (1959)
- * Lackey, J. B., Hendrickson, E. R., Biochemical Bases of Anaerobic Digestion, in Biological Treatment of Sewage and Industrial Wastes, Volume 2, EDS. J. McCabe, and W. W. Eckenfelter, Jr., Reinhold Publishing Corporation, New York (1958) 9-24
- * Leone, D. E., Contract NASw-95, U413-63-041, General Dynamics, Electric Boat Division, Groton, Conn. (1963)
- * McKinney, R. E., Microbiology for Sanitary Engineers, McGraw-Hill Book Co., Inc., New York, 1962
- * McKinney, R. E., and R. G. Weichlein, Appl. Microbiol. 1, 259 (1953)
- * Melek, I., Development and Further Perspective of the Continuous Cultivation of Microorganisms, p. 3-20, In Continuous Culture of Microorganisms, Society of Chemical Industry, London, 1961
- * Moser, H., The Dynamics of Bacterial Populations Maintained in the Chemostat, Carnegie Institute of Washington, Washington, D.C., 1958
- * Moyer, J. E., Aerobic Waste Disposal Systems, in Biologistics for Space Systems Symposium (May 1962), Technical Documentary Report No. AMRL-TDR-62-116 (October 1962), 281-289
- * Moyer, J. E., TDR No. AMRL-TDR-62-116, Aerospace Medical Division, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio, p. 281 (1962)
- * Piper, W. O., and Quon, J. E., Growth of Chlorella on Products from the Incineration of Human Wastes, in Biologistics for Space Systems Symposium (May 1962), Technical Documentary Report No. AMRL-TDR-62-116 (October 1962), 315-327
- * Pote, H. L., Waste Conversion by Anaerobic Thermophilic Methods, in Biologistics for Space Systems Symposium (May 1962), Technical Documentary Report No. AMRL-TDR-62-116 (October 1962), 290-294

- * Pote, H. L., Gafford, R. D., and Richardson, D. E., Anaerobic Waste Processing for Closed Ecological Systems, Advances in Astronautical Sciences, 6 (1961), 399-403
- * Rogovskaya, Ts. I. and M. F. Lazareva, Mikrobiologika, 28, 565 (1959)
- * Rosebury, T., Microorganisms Indigenous to Man, McGraw-Hill Book Co., Inc., New York, 1962, p. 435
- * Ruckhoft, C. C., J. G. Kallas and G. P. Edwards, J. Bacteriol., 19, 269 (1930)
- * Smith, H. W. and W. E. Crabb, J. Pathol. Bacteriol., 82, 53 (1961)
- Mississippi State U., State College, Biochemical Study of Mixed Culture Prototype in a Closed Ecological System, NASA
- * Progress Report No. VI, Dec. 1, 1962 - July 1, 1963, Tischer, R. G., 1963 34p 10 refs /NASA Grant NSG-80-60/ /NASA CR-50860/ OTS- \$3.60 PH, \$1.22 MF N63-20248
- * Tuhrman, R. E., Treating Waste Waters for Cities and Industries p. 647, in Water, The Yearbook of Agriculture, U.S. Dept. of Agriculture (1955).
- * Zeff, J. D., Bambenek, R. A., Development of a Unit for Recovery of Water and Disposal or Storage of Solids from Human Wastes, Part I The Study Phase, Wright Air Development Center Technical Report 58-562 (1959)

11A2 PRODUCTION OF AMMONIA AND HYDROGEN

- * Alexander, M., Introduction to Soil Microbiology, p. 259, John Wiley and Sons, inc., N.Y. (1961)
- * Cohen, J. S. and Burris, 1955, A Method for the Culture of Hydrogen Bacteria, J. Bacteriol., 69, 316-319
- * Cooke, J. V. and H. R. Keith, J. Bacteriol., 13, 315 (1927)
- * Cowles, P. B. and L. F. Rettger, J. Bacteriol., 21, 167 (1931)
- * Fuller, W. H. and A. G. Norman, J. Bacteriol., 46, 273 (1943)
- * Fuller, W. H. and A. G. Norman, J. Bacteriol., 46, 281 (1943)
- * Fuller, W. H. and A. G. Norman, J. Bacteriol., 46, 291 (1943)

- * Gest, H., Bacteriol. Revs., 18, 43 (1954)
- * Gibbons, R. J. and R. N. Doetsch, J. Bacteriol., 77, 417 (1959)
- * Hermann, E. R., J. Soc. Engineering Division Proc. American Soc. Civil Engineers, 88, SA 5, 1, (1962)
- * Hungate, R. E., Bact. Revs., 14, 1 (1950)
- * Khouvine, Y., Ann. de l' Inst. Past., 37, 711 (1923)
- Melpar Corp., Contract DA 36-039 SC-90878, Second Quarterly Progress Report, 1 Oct. - 31 Dec. 1962 (1963)
- * Nagliski, J., J. W. White, Jr., S. R. Hoover and J. J. Willamin, J. Bacteriol., 49, 563(1945)
- * Nisman, R., M. Raynard, and G. N. Cohen, Arch. Biochem., 16, 473 (1948)
- Joint Publications Research Service, Washington, D. C., New Developments in Biochemistry. S. Severin. June 15, 1962. 12p Transl. from Pravda/Moscow/, Sept. 16, 1961. p. 6. /JPRS-14129/ Distributed by OTS. N62-13101
- * Stickland, L. H., Bioch. J., 28, 1746 (1934)
- * Wheaton, R. B., J. J. Symons, N. G. Roth and H. H. Morris, TDR No. AMRL-TDR-62-116, Aerospace Medical Division, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio, p. 295 (1962)
- * Wiley, W. H. and J. R. Stokes, J. Bacteriol., 84, 730 (1962)
- * Zobell, C. E., Microbial Transformation of Molecular Hydrogen in Marine Sediments with Particular Reference to Petroleum, Am. Assoc. Petroleum Geol. 31, 1709 (1947)
- * Zobell, C. E., Assimilation of Hydrocarbons by Microorganisms, Advances in Enzymology 10, 443 (1950)

IIB ENZYMES

1131 CELLULASES

- * Basu and Whitaker, Arch. Biochem. Biophys., 42, 12 (1953)
- * Duncan, Manners and Ross, Biochem. J., 63, 44 (1956)

- * Studies in Cellulose Decomposition, Enebo, Stockholm (1954)
- * Enebo et. al., J. of the Inst. of Brew., 59, 207 (1953)
- * Festenstein, Biochem. J., 69, 562 (1958)
- * Biological Degradation of Cellulose, J. A. Gascoigne and M. M. Casoigne, Butterworths, London, England (1960)
- * Greenfield and Lane, J. Biol. Chem., 204, 669(1953)
- * Holo and Szilagyi, Ind. agr. et. aliment. (Paris) 74, 131 (1957)
- * Hungate, Biol. Bull., 83, 303(1942)
- * Husemann and Lotterle, Makromol. Chem., 4, 278 (1950)
- * Jermyn, Aust. J. Sci. Res., B5, 409 (1952)
- * Karrer and Illing, Kolloidzschr., 36, 91 (1925)
- * Koviman, Enzymologia, 18, 22 (1957)
- * Misra and Ranganathan, Proc. Ind. Acad. Sci., 39B, 100 (1954)
- * Nisizawa, J. Biochem. (Japan) 42, 825-827 (1955)
- * Okamoto and Asai, J. Agric, Chem. Soc. (Japan), 26 (1952)
- * Reese, Gilligan and Norkraus, Physiol. Plant, 5, 379 (1952)
- * Saunders, Sire and Genest, J. Biol. Chem., 174, 697 (1948)
- * Seillere, C. R., Soc. Bio. (Paris), 61, 205 (1906)
- * Seillere, C. R., Soc. Bio. (Paris), 63, 151 (1907)
- * Seillere, C. R., Soc. Bio. (Paris), 68, 107 (1910)
- * Sison, Schubert and Nord, Arch. Biochem. Biophys., 75, 260 (1958)
- * Stone, Ph.D. Thesis, University of London (1954)
- * Stone, Biochem. J., 66, 1 (1957)
- * Toyama, J. Ferment. Technol. (Japan), 34, 281 (1956)

- * Whitaker, Arch. Biochem. Biophys., 43, 253 (1953)
- * Whitaker, Science, 116, 90 (1952)

IIB2 PECTINESTERASES

- * Advances in Enzymology, Vol 20, H. Deuel and E. Stutz, Interscience Publishers, Inc., N.Y. edited by F. F. Nord, (1958)
- * Holden, M., Biochem. J., 40, 103 (1946)
- * Lineweaver, H., and G. A. Ballow, Arch. Biochem., 6, 373 (1945)
- * Advances in Enzymology, Vol. 11, H. Lineweaver and E. F. Jansen, Interscience Publishers, Inc., N. Y., edited by F. F. Nord (1951)
- * MacDonnell, L. R., E. F. Jansen and H. Lineweaver, Arch. Biochem., 6, 389 (1945)
- * McCulloch, R. J., J. C. Moyer and Z. I. Kertesz, Arch. Biochem., 10, 479 (1946)
- * McCulloch, R. J., Z. I. Kertesz, Arch. Biochem., 13, 217 (1947)
- * Mills, C. B., Biochem. J., 44, 302 (1949)
- * Owens, H. S., R. M. McCready and W. D. Maclay, Ind. Eng. Chem. 36, 936 (1944)
- * Pithawala, R. R., C. R. Savur and A. Screenivasan, Arch. Biochem., 17, 235 (1948)
- * Schultz, T. H., H. Lotzkar, H. S. Owens and W. D. Maclay, J. Phys. Chem., 49, 554 (1945)

IIB3 POLYGALACTURASES

- * Deuel, H., and F. Weber, Helv. Chim. Acta, 29, 1872 (1946)
- * Jansen, E. F., L. R. MacDonnell and R. Jang, Arch. Biochem. 8, 113 (1945)
- * Jansen, E. F., R. Jang, L. R. MacDonnell, Arch. Biochem. 15, 415 (1947)
- * Koch, J., Fruchtsaft-Ind., 1, 66 (1956)
- * Lineweaver, H., R. Jang and E. F. Jansen, Arch. Biochem., 20, 131 (1949)

- * Ozawa, J., Ber. Ohara Inst. landwirtsch. Forsch. Kuraskiki Japan, 9, 431 (1951)
- * Pallman, H. and H. Deuel, H. Chimia, 1, 27 (1947)
- * Schubert, E., Biochem. J., 78, 323 (1952)
- * Schubert, E., Melliand Textilber, 8, (1954)

IIB4 LIPASES

- * Bayliss, M., D. Glick and R. A. Siem, J. Bacteriol., 55, 307 (1948)
- * Biochemical Problems of Lipids, B. Borgstrom, G. Popjak and E. LeBreton, eds., Butterworths, London, England (1956), 179
- * Methods in Enzymology, Vol. 1, edited by Sidney Colowick and Nathan Kaplan, Academic Press Inc., New York (1955)
- * Connstein, W., E. Hoyer and H. von Wartenberg, Ber. 35, 3988 (1902)
- * Constantin, M. J., L. Pasero and P. Desnuelle, Biochem. et. Biophys. Acta, 43, 103 (1960)
- * The Enzymes of Lipid Metabolism, edited by P. Desnuelle, Pergamon Press, London, England (1961)
- * Fiore, J. V. and F. F. Nord, Arch. Biochem., 26, 382 (1950)
- * Goldblith, S. and E. Wicks, ASD Technical Report 61-419, ASTIA 266882 (1961)
- * Gorbach, G. and H. Guntner, Sitzber. Akad. Wiss. Wein. Mathnaturw, Kl, Abt. IIB, 141, 415 (1932)
- * Kimmel, J. R. and E. L. Smith, J. Biol. Chem., 207, 515 (1954)
- * Marchis-Monrew, G., L. Sarda and P. Desnuelle, Biochem. et. Biophys. Acta, 41, 358 (1960)
- * Savory, P. and P. Desnuelle, Biochem. et. Biophys. Acta, 21, 349 (1956)
- * Schonheyder, F. and K. Volqvartz, Enzymologia, 11, 178 (1944)
- * Schonheyder, F. and K. Volqvartz, Acta Physiol. Scand., 10, 62 (1945); 11, 349 (1946)

- * Starr, M. P. and W. H. Burkholder, *Phytopathology*, 32, 598(1942)
- * Weinstein, S. S. and A. M. Wynne, *J. Biol. Chem.*, 112, 641, 649 (1935-36)
- * Wills, E. D., *Biochem. Biophys. Acta*, 40, 487 (1950)
- * Wills, E. D., *Biochem. J.*, 57, 109 (1954)
- * Wills, E. D., *Biochem. J.*, 69, 17 (1958)

11B5 PAPAINS

- * Anson, M. L., *J. Gen. Physiol.* 22, 79 (1938)
- * Ball, A. K. and H. Lineweaver, *J. Biol. Chem.*, 130, 669 (1939)
- * Calvery, H. O., *J. Biol. Chem.*, 102, 73 (1933)
- * Hill, R. L., H. C. Schwartz and E. L. Smith, *J. Biol. Chem.*, 234, 573 (1959)
- * Hwang, K. and A. C. Ewy, *Ann. New York Acad. Sci.*, 54, 161 (1951)
- * Irving, G. W. Jr., J. S. Fruton and M. Bergmann, *J. Biol. Chem.*, 138, 231 (1941)
- * Kinzel, J. R. and E. L. Smith, *J. Biol. Chem.*, 207, 515 (1954)
- * Kunitz, M., *J. Gen. Physiol.*, 30, 311 (1947)
- * Lineweaver, H. and S. Schimmer, *Enzymologia*, 10, 81 (1941)

11B6 UREASES

- * *Methods in Enzymology*, Vol. 2, edited by Sidney P. Colowick and Nathan O. Kaplan, Academic Press, Inc., New York (1955)
- * Dounce, A. L., *J. Biol. Chem.*, 140, 307 (1941)
- * Fosman, G. D. and C. Niemann, *J. Am. Chem. Soc.*, 73, 1646 (1951)
- * Gorin, et al., *Biochemistry*, 1, 911 (1962)
- * Hellerman, L., F. B. Chinard and U. R. Dietz, *J. Biol. Chem.*, 147, 443 (1943)

- * Kistiakowsky, C. B. and R. Lumry, J. Am. Chem. Soc., 71, 2006 (1949)
- * Laidler, K. J. and J. P. Hoarle, J. Am. Chem. Soc., 72, 2489 (1950)
- * Larson, A. D. and R. E. Kallio, J. Bacteriol., 68, 67 (1954)
- * Shaw, W. H. R. and C. B. Kistiakowsky, J. Am. Chem. Soc., 72, 2817 (1950)
- * The Enzymes, 1st Ed., Vol. 1, edited by J. B. Sumner and K. Myrbo^Uck, Academic Press, Inc. New York (1951)
- * Sumner, J. B. and L. O. Poland, Proc. Soc. Exptl. Biol. Med., 30, 553 (1933)
- * Wall, M. C. and K. J. Laidler, ABB, 43, 307 (1953)
- * Wall, M. C. and K. J. Laidler, ABB, 43, 299 (1953)

11B7 SULFATE REDUCTASES

- * Allen, L. A., The Effect of Nitro-Compounds and Some Other Substances on Production of Hydrogen Sulphide by Sulphate-reducing Bacteria in Sewage, Proc. Soc. Appl. Bacteriol. 20, 26-28 (1949)
- * Booth, G. H. and Tiller, 1960, Polarization Studies of Mild Steel in Cultures of Sulphate-reducing Bacteria, Trans. Faraday Soc., 56, 1689-1696
- * Butlin, K. R., Adams, and Thomas, 1949, The Isolation and Cultivation of Sulphate-reducing Bacteria, J. Gen. Microbiol., 3, 46-59
- * Gregory, J. D. and P. W. Robbins, Metabolism of Sulfur Compounds (Sulfate Metabolism), Ann. Rev. Biochem. 29, 347-364 (1960)
- * Lazzarini, R. A., The Reduction of Nitrite by Enzymes of Escherichia Coli, Ph. D. Thesis, University of California, (Los Angeles), (June 1960)
- * Miller, L. P., 1950, Tolerance of Sulfate-reducing Bacteria to Hydrogen Sulfide, Contrib. Boyce Thompson Inst., 16, 73-83
- * Miller, L. P., 1949, Stimulation of Hydrogen Sulfide Production by Sulfate-reducing Bacteria, Contrib. Boyce Thompson Inst., 15, 467-474

- * Postgate, J. R., Sulfate Reduction by Bacteria, Annual Rev. Microbiology 13, 505 (1959)
- * H. R. Schreiner and A. P. Rinfret, Effect of Water-Soluble Polymers on Nitrate Reductase Activity, Nature 190, 1110-1112 (1961)
- * Sisler, F. D. and ZoBell, 1951, Nitrogen Fixation by Sulfate-reducing Bacteria Indicated by Nitrogen/argon Ratios, Science, 113, 511-512
- * Starkey, R. L. Sulfate-reducing Bacteria - Physiology and Practical Significance, U. of Md., Dept. of Microbiology Bulletin, 1960
- * Zobell, C. E., Rittenberg, S. C., Sulfate-reducing Bacteria in Marine Sediments, J. Marine Research (Sears Foundation 7, 602 (1948)
- * ZoBell, C. E., 1958, Ecology of Sulfate-reducing Bacteria, Producers Monthly, 22, 12-29

11B8 HYDROGENASES

- * Adams, M. E., Butlin, Hollands and Postgate, 1951, The Role of Hydrogenase in the Autotrophy of Desulphovibrio, Research, 4, 245-246
- * Kresna, A. I., E. Riklis, and D. Rittenberg, The Purification and Properties of the Hydrogenase of Desulfovibrio desulfuricans J. Biol. Chem. 235, 2717-2720, (1960)
- * Marshall, C. E. and Bergman, W. E. J. Am Chem. Soc. 63, 1911 1941
- * Marshall, C. E. and Krinbil, C. A., J. Am. Chem. Soc. 64, 1814, 1942
- * Mechalias, B. J. and S. C. Rittenberg, Energy Coupling in Desulfovibrio desulfuricans, Bacteriol. Proc., Abstr. P53, 164, (1960)
- * Ormerod, J. G., and H. Gest, Hydrogen Photosynthesis and Alternative Metabolic Pathways in Photosynthetic Bacteria, Bacteriol. Rev. 26, 51-66 (1962)
- * Ricklis, E. and Rittenberg, 1961, Some Observations on the Enzyme, Hydrogenase, J. Biol. Chem, 236, 2526-2529

- * Sadana, J. C. and A. V. Morey, The Purification of Hydrogenase of *Desulfovibrio desulfuricans*, *Biochem. Biophys. Acta.* 32, 592-593, (1959)
- * B. L. Vallee and F. L. Hock, Yeast Alcohol Dehydrogenase, A Zinc Metalloenzyme, *J. Am. Chem. Soc.*, 77, 821 (1955)
- * F. H. Westheimer, H. F. Fisher, E. E. Coon, and B. Vennesland, The Enzymatic Transfer of Hydrogen from Alcohol to DPN, *J. Am. Chem. Soc.*, 73, 2403 (1951)

II B9 CATALASES

- * B. Chance, D. S. Greenstein, and F. J. W. Roughton, The Mechanism of Catalase Action. I. Steady-State Analysis. *Arch. Biochem. Biophys.* 37, 301 (1952)

II B10 ROLE OF METAL IONS IN ENZYME SYSTEMS

- * Lehninger, A. L., Role of Metal Ions in Enzyme Systems, *Physiological Review*, 30, 393-483 (1950). 225 Reference
- * H. Kubo and M. Iwatsubo, A Spectrometric Study of Metals in Some Oxidases, *Bunko Kenkyu* 1, No. 4, p 15-21 (1949)

III BIOELECTROCHEMICAL CONVERTERS AND RELATED STUDIES

IIIA FUNDAMENTAL INVESTIGATIONS

IIIA1 ELECTRODE STUDIES

- * Electrode Kinetics and Fuel Cells. L. G. Austin /Pennsylvania State University, Dept. of Fuel Technology, University Park, Pa./ IEEE, Proceedings, Vol. 51, May 1963, P. 820-837. 15 Refs. A63-16775
- * Booth, G. H., and Tiller, A. K., *Faraday Society*, 1960, 56, 1689
- * Booth, G. H., and Wormwell, F., *First International Congress on Metallic Corrosion*, 1961, p. 341
- Aeronautronic, Newport Beach, Calif. Study of Basic Bio-Electrochemistry Quarterly Engineering Progress Report, 19 Mar. - 19 June 1963, M. H. Boyer, R. C. Bean, Y. H. Inami, E. R. Walwick, and R. E. Kay July 19, 1963 41p 12 refs /NASA Contrac NASW-655/ /NASA CR-51113, U-2208/ OTS \$4.60 PH, \$1.43 MF, N63-21729

- Aeronautic, Newport Beach, Calif. Study of Basic Bio-Electro-chemistry Quarterly Engineering Progress Report, 20 June. - 19 Sept. 1963, M. H. Boyer, R. C. Bean, Y. H. Inami, and W. C. Shepherd 19 Oct. 1963 69p /NASA Contract NASW-655/ /NASA CR-52607, U-2332/ OTS- \$6.60 PH, \$2.27 MF, N64-11103
- * Magna Corp., Anaheim, Calif. Research and Development Div. Biochemical Fuel Cells Fourth Quarterly Progress Report, Apr. 1 - June 30, 1963, J. M. Brake, W. R. Momyer, and H. P. Silverman 1963 30p 11 refs., /Contract DA 36-039-SC-90866/ N63-23418
- * Magna Corp., Anaheim, Calif. Research and Development Div. Biochemical Fuel Cells, Fifth Quarterly Progress Report, 1 Jul. - 30 Sept. 1963, J. M. Brake, W. R. Momyer, and H. P. Silverman 1963 38p refs., /Contract DA-36-039-SC-90866/ /AD-426937/ N64-13968
- * Magna Corp., Anaheim, Calif. Research and Development Div., Research on Applied Bioelectrochemistry, Quarterly progress Report No. 2, Jul. 1 - Sep. 30, 1963, James H. Canfield, Oct. 1963 46p refs /NASA Contract NASW-623/ /NASA CR-52606, 339/7014/ T6, 714-772-1261/ OTS- \$4.60 PM, \$1.58 MF, N64-11323
- * Carr, C. W. Gregor, H. P., and Sollner, K. J. Gen. Physiol. 28, 179, 1945
- * Clark, W. M., Oxidation-Reduction Potentials of Organic Systems, Williams & Wilkins, Baltimore, 1960
- * Cohen, B., The Bacterial Culture as an Electrical Half-Cell, Jour. of Bacteriology, Vol. 21, p. 18 (1931)
- * Cohen, Barnett, 1931, The Bacterial Culture as an Electrical Half-Cell, J. Bacteriol. 21, 18-19
- * Dolin, M. I., Survey of Microbial Electron Transport Mechanisms Chapter 6 of The Bacteria Vol. II, edited by I. C. Gunsalus and R. Y. Stanier, Academic Press 1961
- * Gatty, O., and Spooner, E. C. R., Electrode Potential Behavior of Corroding Metals in Aqueous Solutions, Oxford, 1938, Chapter X.
- * Gregor, H. P. and Sollner, K., J. Phys. Chem. 58, 409, 1954
- * The Action of Magnetic Field on the Sodium Transport Across the Cell Membrane, T. Gualtierotti and V. Capraro /University of Milan, Institute of Human and General Physiology, Milan, Italy/

Cospar, International Space Science Symposium, 4th, Warsaw
Poland, June 3-11, 1963, Paper. 8p A63-18961

- * Hadley, R. F., 1943, The Influence of *Sporovibrio desulfuricans* on the Current and Potential Behavior of Corroding Iron, Bu. Stand., Soil Corrosion Conf., St. Louis, pp76
- * Kedrov, K. P., The Characteristics of the Oxidation-Reduction Processes in the Intestines, Biochem., J. (Ukraine)11, 243 (1938)
- Union Carbide Corp., Parma, Ohio, Thin Fuel Cell Electrodes
- * Quarterly Report No. 1, 1 June - 1 Sep. 1963, K. V. Kordesch Fort Monmouth, N.J. Army Electron. Res. and Develop. Lab. 1963 167p Refs., /Contract DA-36-039-AMC-02314/E// /AD-425031/ N64-12058
- Joint Publications Research Service, Washington, D.C., Bio-geochemistry of Sulfur, S. I. Kuznetsov 9 Jan. 1964 20p Refs., Transl. into English of an Article from Izv. Akad. Nauk SSSR Ser. Biol. /Moscow/, No. 5, 1963 p 668-680, /JPRS-22672, OTS-64-21249/ OTS- \$0.50, N64-13007
- * Lemaire, R., On the Oxidation-Reduction Potential of Arterial Blood in Vivo, Soc. de Biologie, Paris, Comptes Rend. 141, 775, (1947)
- * Littlewood, D. and Postgate, 1957, On the Osmotic Behaviour of *Desulphovibrio desulphuricans*, J. Gen. Microbiol., 17, 596-603
- Magna Products, Inc., Bio-Electrode Ocean Batter Research and Development Program, Tech. Rpt., July 1961, 9
- * Mechalias, B. and Rittenberg, 1960, Energy Coupling in *Desulfovibrio desulfuricans*, J. Bacteriol., 80, 501-507
- * Nagamatsu, M., Seiyania, T., and Sakai, W., J. Electrochem. Soc. Japan 21, 270-4, 1953
- * Postgate, J., Progress in Industrial Microbiology, Vol. 2, Interscience Publishers, Inc., New York (1960)
- * Potter, M. C., Electrical Effects Accompanying the Decomposition of Organic Compounds Proc. Roy. Soc. (London) Series B Vol. 84 (1911)

- Magna Corp. Research and Development Labs., Anaheim, Calif.
- * Biochemical Fuel Cells, G. H. Rohrback, W. R. Scott, and J. H. Canfield in Army Signal Research and Development Lab. Electronic Components Dept., Sources Conf., Fort Monmouth, N.J. May 22-24, 1962 p18-21, /See N63-14123 09-06/ /Contracts NOBS-78659, NOBS-84243, and NOBS-84622/ N63-14130
 - * Ross, S., Markarian, M., and Schroeder, W., U. S. Patent 2, 648, 717
 - * Sawada, T., Oshima, T., Suzuki, I., Oxidation-Reduction Potentials in Relation to the Cultivation of Entamoeba Histolytica and to the Establishment of Amoebic Infection. Guma J. of Med. Sciences (Japan) 2, 127-35 (1953)
 - * Sisler, F. D. and Zobell, C. E., J. Bact., 60, 1950, 747
 - * Sollner, K. and Gregor, H. P., J. Phys. Chem. 51, 299, 1947
 - * Sollner, K. J. Electrochem. Soc. 97, 139C, 1950
 - * Sollner, K. and Gregor, H. P., J. Coll. Sci. 6, 557, 1951
 - * Sollner, K. and Gregor, H. P., J. Coll. Sci. 7, 37, 1952
 - * Tiller, A. K., and Booth, G. H., Trans. Faraday Society, Part II, 1962, 58, 110
 - * Trivedi, A. K. M. and Divatia, A. S., Proc. Indian Acad. Sci. 37A 33-37, 1953
- General Dynamics/Fort Worth, Tex. Investigation of Electro-Chemical Properties of Liquid and Frozen Electrolytes at Low Temperatures, H. J. Weltman, 4 Nov. 1963 65p Refs., /Contract AF 33/657/-11214/ /FGT-2917, AD-423614/, N64-12318
- * Joint Publications Research Service, Washington, D.C., Studies in Biochemistry, N. I. Yelkina et al 2 Jul. 1963 25p Refs., Transl. into English of Three Articles from Vopr. Med. Khim. /Moscow/, V.9, No. 2, 1963 p 154-160, 180-188 /JPRS-19995 OTS-63-31185/ OTS- \$0.75, N64-12570

IIIA2 BIOELECTRIC ACTIVITY

- * Abrams, I. and Sollner, K., J. Gen. Physiol. 26, 369, 1943
- * Bennett, M.V.L. and Wurzel, M. and Grundfest, H., The Electrophysiology of Electric Organs of Marine Electric Fishes I, J. Gen. Physiol., 44, 757 (1961)
- * Bennett, M.V.L. and Grundfest, H., The Electrophysiology of Electric Organs of Marine Electric Fishes II, J. Gen. Physiol. 44, 805 (1961)
- * Bennett, M.V.L. and Grundfest, H., The Electrophysiology of Electric Organs of Marine Electric Fishes III, J. Gen. Physiol. 44, 819 (1961)
- * Bennett, M.V.L. and Grundfest, H. and Keynes, R. D., The Discharge Mechanisms of the Electric Catfish, J. Physiol., 143, 52 (1958)
- * Bennett, M.V.L. and Grundfest, H., Electrophysiology of Electric Organ in Gymnotus Carpo, J. Gen. Physiol., 42, 1067 (1959)
- * Carr, C. W. and Sollner, K. J. Gen. Physiol. 28, 119, 1944
- * Chance, Britton, Free Radicals in Biological Systems, Academic Press, New York (1961)
- * Cox, R. T., Coates, C. W. and Brown, M. V., Electrical Characteristics of Electric Tissue, Ann. N.Y. Acad. Sci. 487-500(1946)
- Naval Radiological Defense Lab., San Francisco, Calif. Sodium
* Dependence of Bioelectric Outputs in Rat Stomach, J. T. Cummins and B. E. Vaughn, Jan. 14, 1963 10p 9 Refs /USNRDL-TR-608/ N63-13573
- California U., Berkeley. Lawrence Radiation Lab. Bioelectric
Sensitivity to Irradiation of the Retina and Visual Pathways,
* C. T. Gaffey and A. K. Kelley, 9 Sep. 1963, 41p refs., /Contract W-7405-ENG-48/ /UCRL-11005/ OTS- \$1.00, N64-11097
- * Grundfest, H., Electric Inexcitability of Synapses and Some Consequences in the Central Nervous System, Physiol. Rev., 37, 337 (1957)
- * Gunsalus and Stanier, The Bacteria, Vol. I: Structure, Academic Press, New York (1960)

- Electroencephalographic Investigation of the Functional State of the Human Central Nervous System Under Conditions of Prolonged Isolation, Elektroentsefalograficheskoe Issledovanie Funktsional'noy Sostoianii Tsentral'noy Nervnoy Sistemy Cheloveka, Nakhodiashchegosya v Usloviyakh Dlitel'noy Odinochestva, G. V. Izosimov and V. I. Miasnikov. Iskusstvennye Sputniki Zemli, No. 15, 1963, p. 120-123. In Russian. A63-18209
- * Johnels, A. F., On the Origin of the Electric Organ in Malapterurus Electricus, Quart. J. Micro. Sci., 97, 455-64 (1956)
- * Kellenberger, K., Microbial Genetics, Cambridge Univ. Press, London (1960)
- * Keynes, R. D., Electric Organs in the Physiology of Fishes, Vol. II, M.E. Brown, ed., Academic Press, Inc., New York (1957) pages 323-43
- Joint Publications Research Service, Washington, D. C. Respiration and Conversion of Energy in the Living Cell, Kotelnikova In its Studies in Biophys. 23 Apr. 1963 p 21-33 /See N64-10795 OTS- \$1.00, N64-10797
- * Langmuir, I. and Waugh, D. F., J. Gen. Physiol., 21, 745-55 (1938)
- * Lessman, H. W., On the Function and Evolution of Electric Organs in Fish, J. Exp. Biol., 35, 156 (1958)
- * Machin, K. E. and Lessman, H. W., The Mode of Operation of the Electric Receptors in Gymnarchus Nitoticus, J. Exp. Biol., 39, 801-811 (1960)
- * Michaelis, L. and Weech, A. W., J. Gen. Physiol. 11, 147, 1927
- * Nachmansohn, D. et al, J. Biol. Chem. 165, 223 (1946)
- Joint Publications Research Service, Washington, D.C., Thermodynamics of Irreversible Processes and the Problems of Biogenesis /Termodinamika Neobratimyykh Protssessov i Problemy Biogeneza/. L. A. Nikolaev. Apr. 2, 1962, 20p. 12 refs., /JPRS-13286/ Transl. from Zhur. Fiz. Khim./Moscow/ V. 36, No. 1, 1962 p. 3-14. Distributed by OTS. N62-12261
- * Robertson, J. David, Progress in Biophysics, Vol. 10, Pergamon Press, New York (1960)
- * Schoffeniels, E., Les Bases Physiques et Chimiques des Potentials Bioelectriques chez Electrophorus Electricus L., Arch. Internat.

Physiol. Biochem., 68, 1-151

- * Schwan, H., Advances in Biological and Medical Physics, Vol. 5, Academic Press, New York (1957)

Dielektricheskaia Pronitsaemost' Biologicheskikh Ob'ektov Dielectric Permeability of Biological Objects. B. I. Sedunov and

- * D. A. Frank-Kamenetskii. Uspekhi Fizicheskikh Nauk, Vol. 79, Apr. 1963, p. 617-639. 86 Refs. In Russian. A63-16889
- * Dielectric Constants of Biological Objects. B. I. Sedunov and D. A. Frank-Kamenetskii. /Uspekhi Fizicheskikh Nauk, Vol 79, Apr. 1963, p. 617-639./ Soviet Physics - Uspekhi, Vol. 6, Sept. Oct. 1963, p. 279-293. 86 Refs. Translation. for Abstract see Accession no. A63-16889 13-16. A63-25129
- * Electrophytogram of Response Rhythms of Plants to Pulsed-Light Stimulation. Aleksandra D. Semenenko, M. A. Khvedelidze, and Agnessa D. Semenenko/Academy of Sciences, Institute of Cybernetics, Tbilisi, Georgian SSR/. /Adademiia Nauk SSSR, Doklady, Vol. 147, Nov. 1962, p. 727-730./ Soviet Physics - Doklady, Vol. 7, May 1963, p. 1061-1063. Translation. A63-17930
- * Sollner, K., Dray, S., Grim, E., and Neihof, R., Electrochemical Studies with Model Membranes, in J. T. Clarke, Ion Transport Across Membranes, Academic Press, New York, (1954), Page 144
- * Suckling, E. E., Bioelectricity, McGraw-Hill, 1961
- Naval Radiological Defense Lab., San Francisco, Calif. Radio-sensitivity of Bioelectric Functions of Rat Stomach and Caecum
- * B. E. Vaughan and A. K. Davis Jan. 8, 1963 28p 24refs, /USNRDL-TR-611/ N63-13079
- * von Wolzogen Zuhr, C. A. H. and Van der Vlugt, L. S., Water (The Hague), 1934, 16, 147
- Aerospace Medical Div. School of Aerospace Medicine, Brooks AFB Tex. Effects of Calcium Deficiency and Excess on Transmembrane
- * Potentials in Frog Heart Technical Documentary Report F. Ware, Aug. 1962 12p Refs., /SAM-TDR-62-100/ N63-85914

IIIA3 ORGANIC MEMBRANES

- * Bodamer, G. W., U.S. Patents 2, 681,319, and 2, 681,320 (June 15, 1954)
- * Clarke, J. T., Marinsky, J. A., Juda, W., Rosenberg, N. W., and Alexander, S., J. Phys. Chem. 56, 100, 1952

- * Gottlieb, M. H., Neihof, R., and Sollner, K., J. Phys. Chem. 61, 154-159, 1957
- * Graydon, W. F. and Stewart, R. J., J. Phys. Chem. 59, 86, 1955
- * Gregor, H. P., and Wetstone, D. M., J. Phys. Chem. 61, 147-151, 1957
- * Gregor, H. P. Jacobson, H., Shair, R. C., and Wetstone, D. M., J. Phys. Chem. 61, 191-197, 1957
- * Hills, G., Kitchener, J., Ovenden, P., Trans. Faraday Soc. 51, 719-728, 1955
- * Juda, W., Rosenberg, N. W., Marinsky, J. A., and Kasper, A. A., J. Am. Chem. Soc. 74, 3736, 1952
- * Juda, W. and McRae, W. A., J. Am. Chem. Soc. 72, 1044, 1950
- * Kosaka, Y. and Tajima, A. J. Chem. Soc. Japan, Ind. Chem. Sect. 56, 279-281, 1953
- * Kressman, T. R. E., Nature 165, 568, 1950
- * Krishnaswamy, N., J. Sci. Ind. Research India 13B, 722-726, 1954
- * Neihof, R., J. Phys. Chem. 58, 916, 1954
- * Peterson, M. and Gregor, H. P., J. Electrochem. Soc. 106, 1051-1061, 1959
- * Sollner, K., and R., Neihof, Arch. Biochem. Biophys. 33, 166, 1951
- * Spinner, I. H., Circie, J., and Graydon, W. F., Can. J. Chem. 32, 143, 1954
- * Stewart, R. J., and W. F., Graydon, J. Phys. Chem. 61, 164-168, 1957
- * Tajima, S., J. Electrochem. Soc. Japan, 22, 67, 1954
- * Wetstone, D. M. and Gregor, H. P., J. Phys. Chem. 61, 151-154, 1957
- * Winger, A. G., Bodamer, C. W., and Kunin, R., J. Electrochem. Soc. 100, 178, 1953

- * Wyllie, M. R. J. and Patnode, H. W., J. Phys. Chem. 54, 204, 1950
- * Wyllie, M. R. J., and Kannaan, S. L., J. Phys. Chem. 58, 73, 1954

IIIB BIOELECTROCHEMICAL FUEL CELLS

- Harvard U. Graduate School of Business Administration,
* Cambridge, Mass. Fuel Cells -Power for the Future, David R. Adams, et al Fuel Cells Res. Assoc., Cambridge, Mass., Oct. 1960 60p. Refs. Fuel Cell Res. Assoc., Cambridge- \$18.75, N63-81905
- * Anonymous, Proceedings of the Biochemical Fuel Cell Session, Power Information Center Report No. PIC-BAT 209/5, (Nov. 1962) Armed Services Technical Information Agency No. 292, 163
- Joint Publication Research Service, Washington, D.C., Direct Conversion of Chemical Energy into Electrical Energy, V. S.
* Bagotskiy and A. N. Frumkin. Sept. 13, 1962. 34p. 19 Refs., Transl. of Article from Vestnik Akad. Nauk SSSR/Moscow/, V. 32. no. 7, 1962. p. 19-32. /JPRS-15257/ OTS- \$3.60. N62-15581
- Phillips Petroleum Co., Atomic Energy Div., Idaho Falls, Idaho, The Design of a Potentiostat for Electrochemical Research
* R. H. Brown, AEC National Reactor Testing Station, May 1963 40p. 8 Refs., /Contract at/10-1/-205/ /ID0-16852/ OTS- \$1.00 N63-17618
- * Fuel Cells - Progress, Programs, and Problems. Ernst M. Cohn, /NASA, Electrochemical Technology Projects, Washington, D.C. In- 12th International Astronautical Congress, Proceedings, Vol. 1. Washington, D. C., Oct. 1-7, 1961. New York and London, Academic Press, Inc., 1963, p. 438-445. A63-21265
- * Preliminary Biochemical Fuel Cell Investigations. E. L. Colichman, Marquardt Corp., Van Nuys, Calif. IEEE, Proceedings, Vol. 51, May 1963, p. 812-819, 12 Refs. A63-16774
- * Davies, J. B. and Yarbrough, H. F.: Preliminary Experiments on a Microbial Fuel Cell, Science, Vol. 137, pp. 615-616, 24 August 1962
- Marquardt Corp., Van Nuys, Calif. Study of Biochemical Fuel Cells Second Quarterly Progress Report, 1 Aug. 1963 - 31 Oct. 1963. George E. Ellis and Edward E. Sweeney 1963 47P /NASA
* Contract NASW-654/ /NASA CR-52813, Rept. 25105/ OTS- \$4.60 PH. \$1.61 MF, N64-11299

- Marquardt Corp., Van Nuys, Calif. Biochemical Fuel Cells
First Quarterly Progress Report, Mar. 22 July 31, 1963
- * George E. Ellis and Edward E. Sweeney Aug. 10, 1963 59p 38 Refs.
/NASA Contract NASW-654/ /NASA CR-51565, Mr-25093/ OTS- \$5.60
PH, \$1.97 MF, N63-22894
 - * Kressman, T. R. E. and Kitchener, J. A., J. Chem. Soc. 1190,
1949
 - * Long, F. H., Biological Energy as a Power Source for a Phys-
iological Telemetering System, 1962 IRE International Convention,
New York City, March 28, 1962
 - * Magna Products, Inc., New Electrical Generators Utilizing Bio-
logical Processes (Proposal), Oct. 1960
 - RCA Service Co., Patrick AFB, Fla. Missile Test Project Ordnance
 - * - Bio-effects - Fuel, O. B. Rawls, R. J. Stilwell, and B. M.
Mc Donald July 1961 121p Refs., /AFMTC-TR-61-14, Supersedes
AFMTC-TN-59-4, AD-260721/ N63-84327
 - General Electric Co., Schenectady, N.Y., Space Sciences Lab.
 - * Two Bio-electrogenic Systems a Preliminary Report, L. W.
Reynolds and J. J. Konikoff Sept. 1962 25p Refs., /R62SD78/
N63-85664
 - * Rohrback, G. H., W. R. Scott and J. H. Canfield, Biochemical
Fuel Cells, 16th Annual Power Sources Conference of U.S. Army
Signal Research and Development Laboratories, May 22-24, 1962
Atlantic City, New Jersey.
 - * Scott, W. R., G. H. Rohrback and M. G. Del Duca, Biochemical
Fuel Cell Potentialities for Space Flight, American Rocket
Society Space Power Systems Conference, September 25-28, 1962
Santa Monica, California
 - * Shaw, M., Biochemical Fuel Cells, Advanced Technology Center,
Electric Autolite Co., Palo Alto, California (Dec 1962)
 - * Sisler, F. D., Fuel Cell Uses Bacteria to Produce Power,
Missiles and Rockets, April 17, 1961
 - Massachusetts Inst. of Tech., Cambridge Research needs for
 - * Energy Conversion Systems, David C. White, in AIAA 2nd Manned
Space Flight Meeting 1963, p 308-328 Refs /See N63-23201 24-01/
N63-23238

Naval Research Lab., Washington, D. C., Direct Energy Conversion
Literature Abstracts July 1963 183p 1077 Refs., OTS- \$3.00
N63-23603